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PROCEEDINGS OF THE

SYMPOSIUM ON SAFEGUARDS

RESEARCH AND DEVELOPMENT

Held at

ARGONNE NATIONAL LABORATORY

JUNE 26, 1967

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WASH-1076
General, Miscellaneous,
and Progress Reports
(TID-4500)
AEC Research and
Development Report

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

CONFIDENTIAL

H.C. \$ 3.00; MN 65

PROCEEDINGS OF THE SYMPOSIUM ON
SAFEGUARDS RESEARCH AND DEVELOPMENT

Held at

Argonne National Laboratory
June 26, 1967

Chairman

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Director, Office of Safeguards and
Materials Management*
U.S. Atomic Energy Commission

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(Classified Session held June 27, 1967)

*As of July 1, 1967.

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INTRODUCTORY REMARKS

S. Lawroski
Argonne National Laboratory
Argonne, Illinois

Since its establishment more than twenty years ago, Argonne has had the privilege of being host to many, many conferences and symposia which have been sponsored by the Atomic Energy Commission or by the Laboratory. The symposium today and tomorrow is sponsored by the Atomic Energy Commission, and we are especially pleased to be the host.

The subject for this symposium: how to devise international nuclear safeguards against the diversion of nuclear materials to weapons applications, makes this the most important conference or symposium we have ever had here. Therefore, we are particularly pleased and honored to have present today so many distinguished people who have agreed to come here to discuss and contribute their ideas on international nuclear safeguards, which is one of the most important topics and problems of our world today. Gentlemen, welcome to the Laboratory.

I would now like to turn the meeting over to General Crowson, who is the Presiding Chairman of this symposium.

INTRODUCTORY REMARKS

Brig. Gen. D. L. Crowson, USAF (Ret.)
Director, Office of Safeguards and
Materials Management
U.S. Atomic Energy Commission
Washington, D.C.

Thank you. From the looks of the group, and from the welcome that we received, the organization and how the timing is going, it bodes well for a very successful conference.

My problem is that I am sort of schizophrenic at this stage of the ball game. I am still in uniform, but I am also the Chairman of a safeguards group that is being established in Washington. As a result, I am not really, shall we say, in a position to bring you any words of advice. I, too, came here to learn.

Let me welcome you all to this general session, the purposes of which, as you well understand, are to discuss research and development as they pertain to safeguards. The main purpose, and let me encourage you to do this, is to have a free exchange of ideas and to make sure that each of you thinks quite carefully about what the future holds as far as research and development in connection with safeguards are concerned.

Much needs to be done, from what reading I have done in this field and what I have learned from various talks with authorities. I believe that a symposium of this sort will get us off on the right foot. We want you to go away from here basically with the ideas that you can contribute and you will contribute.

The first session of which I am acting as Chairman this morning covers the general background and the general information concerning both national and international safeguards. Many of you have raised questions about the problems of how the AEC intends to organize, and what its responsibilities are. We have added an additional speaker to the agenda: John Vinciguerra, who will cover the present organization of the AEC and what is expected of it as far as the new organization is concerned, and basically what my responsibilities will be, those of the present nuclear materials management people, as well as the International Affairs people.

As I said, the first session is to cover the basic background in the international safeguards area, and we are very privileged and honored this morning to have as our keynote speaker the Chairman of the Atomic Energy Commission, Dr. Seaborg.

NEW PROBLEMS IN SAFEGUARDING THE ATOM

Dr. Glenn T. Seaborg, Chairman
U.S. Atomic Energy Commission
Washington, D.C.

I am pleased to have the opportunity to introduce the important subject of this symposium. Naturally, I am gratified to see so many outstanding people here from the scientific community, university and contractor laboratories, and Government agencies concerned with national and international security.

For some years, the Atomic Energy Commission has sponsored technical research and development directed toward finding new and more efficient methods to ensure that fissionable materials will not be diverted for unauthorized purposes or that such diversions will be detected. Today we are meeting near the same site where a generation ago the talents and efforts of great minds released the same nuclear genie that we are now seeking to control under more effective safeguards. And today, as we stand at a crossroad in the evolution of man's control of the atom, we are again calling upon the talents and efforts of great minds to meet new challenges.

We have been successful in controlling the atom from a technical standpoint and in harnessing its power to the benefit of mankind. We are now faced with the necessity to ensure that this vast source of energy is directed solely toward fulfilling man's hopes for peace and progress. The application of safeguards against the unauthorized diversion of special nuclear material has been a fundamental element of United States policy, both domestically and internationally. The urgency and importance attached to safeguards have been repeatedly stressed by our President and leaders at the highest levels in both the executive and legislative branches of Government. The same urgency and importance are also reflected by the charter of the International Atomic Energy Agency and the current negotiations for a nonproliferation treaty. And I would like to emphasize that our safeguards program has benefited greatly from the enlightened initiative and support of the Joint Committee on Atomic Energy.

Since 1958 the Atomic Energy Commission has conducted nearly 700 safeguards inspections in 29 foreign countries. Euratom has carried out more than 650 inspections in its six member states. The International Atomic Energy Agency is safeguarding more than 50 facilities in 23 countries. Forty countries have agreed to open some or all of their peaceful nuclear activities to safeguards. The United States is now in the process of transferring its international safeguards to the International Atomic Energy Agency.

The problem of safeguards becomes more serious and more complex as the peaceful applications of nuclear energy expand. One reason is that the rapid rate of growth of nuclear power will stimulate a tremendous increase in the worldwide production of plutonium. Projections indicate that by 1980 there will be enough plutonium produced around the globe per day for more than 100,000 megawatt days of electric power--or for tons of nuclear weapons. This is an enormous potential for both good and evil. Another reason is that in the United States the Government's role in the ownership and management of special nuclear material is rapidly changing so that in the future large quantities of special nuclear material will be owned by private enterprise. These changing conditions call for new methods to strengthen safeguards programs at both the international and domestic levels.

Our domestic safeguards program is closely related to our efforts toward more effective international safeguards. In our domestic program, since 1947 we have conducted over 2000 inspections. Concerning the necessity for domestic safeguards, the Government certainly recognizes the integrity of responsible business organizations. It is possible, however, that because of the high strategic value of special nuclear material, black-market problems may emerge and criminal elements may seek to acquire it. Foreign interests, too, may seek unauthorized access. Our objective domestically should be to build a strong safeguards system that will contribute both experience and technology to a more effective international safeguards system.

To meet changes in the conditions affecting safeguards, the Atomic Energy Commission has recently taken a number of significant steps to strengthen its safeguards programs domestically. I would like to take a few minutes to highlight some of these actions.

First, the AEC is making a thorough evaluation of its domestic policies and controls for special nuclear material. We are working to identify areas where improvements could be made to ensure that fissionable material is properly accounted for in the interests of national security.

As part of this evaluation, the Commission appointed a special ad hoc Advisory Panel on Safeguarding Special Nuclear Material. The Panel, composed of persons outside the Government, was asked to assess the effectiveness of existing safeguards policies and procedures, and to recommend any improvement that the Panel believed should be made. Under the chairmanship of Dr. Ralph F. Lumb, this Panel submitted its report, which was released last month. Some of its recommendations have already been adopted by the AEC, and others are now being reviewed.

Second, the AEC has amended its domestic regulations for the protection of fissionable material. Legislation passed in 1964 permits

private ownership of plutonium, uranium-233, and uranium enriched in the isotope 235 that is not subject to accountability controls exercised by the AEC through contractual relationships. Earlier this year we amended our regulations concerning the responsibilities of licensees to control and account for special nuclear material. In essence, the new regulations codify certain accountability requirements that have been imposed on licensees with respect to Government-owned special nuclear material and make them applicable to material that is privately owned. This is a step that is essential to keep safeguards up to date at a time when the growth of peaceful nuclear applications is resulting in the private ownership of fissionable material in increasing amounts.

Third, we have strengthened our organization for carrying out safeguards. We have established a new Office of Safeguards and Materials Management that will report directly to the General Manager, and have overall responsibility for domestic and international safeguards and for inspection programs. This Office will be the focal point for developing and coordinating the AEC's safeguards policies and programs among other AEC divisions and offices and other Government agencies. Its first Director will be General Delmar Crowson, the chairman of this symposium. Del is retiring from the Air Force after a distinguished military career, which included important assignments to nuclear activities and research and development work. Since 1962 he has done an outstanding job as Deputy Director and then Director of the AEC's Division of Military Application, and we are fortunate that he is going to stay with us in his new capacity.

The AEC has also created a new Division of Nuclear Materials Safeguards under the Director of Regulation. This Division will administer safeguards for special nuclear material held by AEC licensees.

The last organizational change I would like to mention is the establishment of an Advisory Committee on Nuclear Materials Safeguards to advise the Commission on matters related to the development of safeguards standards.

Fourth, another safeguards innovation is the plan to station AEC resident inspectors at four private facilities that process significant quantities of special nuclear material. These facilities are the Nuclear Fuel Services, Inc. chemical processing plant at West Valley, New York; the fuel-fabrication facility of Nuclear Fuel Services, Inc. at Erwin, Tennessee; the fuel-fabrication plants of Nuclear Materials and Equipment Corporation at Apollo and Leechburg, Pennsylvania; and United Nuclear Corporation's fuel-fabrication plant at Hematite, Missouri. All of these facilities handle highly enriched uranium or plutonium.

Resident inspection will give us valuable experience toward the development of more effective and efficient safeguards practices. The results should be useful to both the United States and the world at large. After a reasonable period of operation, estimated at about one year, the program will be evaluated by the Commission to determine what arrangements should be made in the future.

Finally, the AEC is increasing its research and development efforts on safeguards techniques to help bridge the gaps between existing knowledge and the technology required for adequate control in the future. In 1966 we spent \$230,000 for research on safeguards; in 1967 we expect to spend about \$1,000,000.

Since 1956 the AEC has spent \$2,400,000 for research specifically on safeguards. Work under this category included studies of inventory control, inspection procedures, monitoring devices, sealing devices, analytical instruments, and new methods for the nondestructive detection, identification, and analysis of individual fission isotopes. During this time an additional \$3,200,000 worth of research was significantly related to safeguards, including work on the development of isotopic and chemical standards for plutonium and uranium, as well as studies to develop and evaluate more sophisticated techniques for analyzing, measuring, and calculating plutonium and uranium when these materials are transferred between facilities or burned in reactors. A summary of the results of research and development contracts awarded by the Atomic Energy Commission and the International Atomic Energy Agency is contained in a booklet entitled Research and Development for Safeguards, which was prepared by the AEC's Division of International Affairs and is dated May 1, 1967.

In the area of research and development, a major problem now facing us is the need for radically new techniques or the refinement of old ones to meet the new conditions. The material control procedures that have worked effectively in the past may not be entirely adequate when special nuclear material is available in larger amounts and more widely distributed, because present measurement uncertainties are too large to guarantee that significant quantities of fissionable material could not be diverted without detection. Among the things we must do is to improve our techniques for ensuring that the material balance in a nuclear fuel cycle can be established with enough precision and accuracy to ascertain whether strategic quantities of nuclear material may or may not have been diverted to unauthorized uses.

I believe that recent advances in computers, solid-state sciences, and instrumentation might be applied to improve methods of verification, including automated instrumentation, that will strengthen safeguards operations. Such methods might enable us to make more accurate measurements of materials in the fuel cycle more efficiently and less

obtrusively. Improved methods of verification, such as automated instrumentation, might also help to minimize the need for periodic on-site inspections and hold down the costs and manpower requirements of safeguards. Moreover, they might become a valuable source of information for plant operators to learn more about what is happening in their facilities.

The safeguard techniques that have been developed up to now have worked effectively without being unduly burdensome. Experience has demonstrated that there is no reason for countries to fear that safeguards must hinder peaceful nuclear progress. But as nuclear industry continues to grow and the requirements for safeguards grow correspondingly, research, development, and future technological innovation may become increasingly important to preserving a safeguards system that is not unduly burdensome. In the future, improved methods of verification could make safeguard operations more satisfactory to all parties both here and abroad. By lessening the requirements for access by inspecting personnel to equipment and facilities that use or produce fissionable material, automated instrumentation could assist in further reducing the largely ungrounded objections to the application of safeguards. In short, I think that developments in automated instrumentation might help to further foster the climate of confidence that is essential to the successful application of safeguards on a worldwide basis.

We also need to develop better means of physical security against theft and unauthorized diversion of special nuclear material. Such means would include, for example, the use of electronic and mechanical codes, seals, and alarms that are essentially tamper-proof. A sophisticated use of these barriers would not only strengthen safeguards, it would also protect the financial interest that governments and industries have in the security of expensive nuclear material.

The problem of physical security over special nuclear material reminds me of our previous success in developing the permissive action links (or PALs) for the physical security of our nuclear weapons deployed within the NATO system. It was a program of research and development carried out by AEC weapons laboratories, strongly supported by the White House and the Joint Committee on Atomic Energy, and pushed with great vigor and ingenuity in 1961 that found a way to put special control devices, or safety locks, on nuclear weapons. The further development of physical security techniques for safeguarding special nuclear material would give us significantly stronger assurance that diversions for nonpeaceful purposes would be detected. If we could have the same degree of assurance that fissionable material is not being clandestinely diverted as we already have that our nuclear weapons will not be used illegally, it would be a great advance for the cause of world peace and security.

It is to meet the challenge of new problems in safeguarding the atom that I am asking for the participation and cooperation of each of you. I earnestly hope that you will give us an outpouring of ideas that will enable us to embark on an aggressive research and development program for safeguards. I recognize that some of the people here have for many years made valuable contributions to the development of safeguards techniques. The agenda for this meeting speaks well for the significant strides being made by those already dedicated to the vital task of developing a more effective safeguards system. I am hopeful that this meeting will swell the ranks of those who are already so dedicated.

GENERAL CROWSON: Thank you very much, Dr. Seaborg, for giving us some objectives at which the conference can shoot.

So long as we are talking about the Atomic Energy Commission, I would like to have you all recognize the importance that the Commissioners themselves place on the safeguards symposium.

Our next speaker is Dr. Smyth, of the International Atomic Energy Agency, whose subject is "Background of International Safeguards." Dr. Smyth carries the rank of Ambassador, but he says he prefers to be called Doctor, because he just can't get the oldtimers in the State Department to recognize his new status.

BACKGROUND OF INTERNATIONAL SAFEGUARDS

H. D. Smyth
U.S. Representative to the International Atomic Energy Agency
Princeton University
Princeton, New Jersey

Gentlemen:

If General Crowson will forgive me, I will correct his introduction a little bit in the interest of clarity. I am the U.S. representative to the International Atomic Energy Agency, with the personal rank of Ambassador. I am employed by the Government of the United States, not by the International Atomic Energy Agency.

This morning I will sketch some of the history of efforts to control the spread of nuclear weapons. Then I will talk about the present situation in international safeguards, and about the differences between the problems of international safeguards and those of domestic safeguards. Such differences are important.

The problem of control of nuclear energy, both nuclear weapons and the peaceful uses of nuclear energy, is an old one. In fact, I had the privilege of serving under Dr. Tolman in the last year or so of World War II on a committee that was investigating these problems. I think we recognized quite clearly then that such problems were going to be serious, but at that time we were able to do little more than define them.

You will recall, or some of you will, that shortly after the war, the United States made a very ambitious effort at international control, starting from the so-called Acheson-Lilienthal Report which considered the problem, and going into the Baruch Plan, which was discussed by the United Nations over a period of several years. That plan in effect suggested international ownership, development, and control of nuclear energy. The Baruch Plan was probably too idealistic for any period, certainly for that period, and it came to nothing, although I find in reading over those old documents that there is much in them that is still of interest and even suggestive.

After the Baruch Plan failed, nothing very much happened for several years. In 1954 the so-called Atoms for Peace Plan was advanced, and we initiated a series of international conferences in Geneva to spread the knowledge of nuclear energy around the world, that is, as much of that knowledge as we could consider was declassifiable at that time.

Let me remind you that there really were four possible courses of action that might have been considered after the war.

One possibility was to say that nuclear energy was such a potential danger that no development of peaceful uses was worth the hazards that the spread of the technology might cause. In other words, we could have just sat on our information and we could have said "Well, we know how to make nuclear weapons; we will keep on making nuclear weapons. Nuclear power is a nice possibility, but we can always burn coal and oil. We don't have to have nuclear power, so why should we run the risk of developing it."

A second possibility would have been a completely secret development. Such a course was advocated by quite a number of people, but had obvious difficulties, at least in a country like ours, a country that is an open society and that we have every intention of maintaining as an open society.

A third possibility was wide-open development, that is, treating this field like any other technological field, giving out all the information that was available, except possibly for patent and trade secret information, and letting the world take care of itself, hoping that other countries would have sense enough either not to make nuclear weapons, or to associate in some way to control them. This possibility was also not chosen.

We in fact chose the fourth possible course of action, wisely I think. We chose to promote the development of atomic energy for peaceful purposes around the world, at the same time establishing a system whereby we hoped we could make sure that any development that we assisted would indeed be limited to peaceful uses, and not diverted in any way to military uses.

This policy resulted in the establishment of what we would call a U.S. safeguards system, which we already had and needed in this country for materials control, and also to some extent for security. Clearly we needed it in other countries to guard against the development of nuclear weapons. This led to the establishment, to which Dr. Seaborg referred, of a system of foreign inspections, so that if we ever sent material or equipment abroad to be used for peaceful purposes, we retained in the treaties that governed the supply of such material the right to send inspectors. So we had in effect a system of U.S. foreign safeguards.

But we believed that this was necessarily a temporary expedient. Even in the middle-Fifties it was felt that eventually some sort of international arrangement was preferable to the U.S. taking the responsibility. I hardly need go into the problems of U.S. inspectors going into foreign countries. Actually, I think in practice such problems have not been serious, but in principle they might well run into difficulties, and it is desirable that such a policing system should be operated by an international organization.

This desire was involved in the establishment of two international organizations in the late Fifties, that is, about ten years ago. One came about for a basically different reason. That was the Euratom organization, which as you know is part of the organization in Western Europe of which the other two parts are the Coal and Steel Community, and the Common Market. The establishment of Euratom at that time has been described to me by some of the Europeans who were involved as being particularly attractive because the Coal and Steel Community and the Common Market were somewhat pedestrian, though obviously desirable from the economic point of view and desirable as a step towards the unification of Western Europe. At that time atomic energy still had the fine romantic appeal that has now been taken over by explorations in outer space.

Evidently, the inspection and safeguarding function of Euratom was a somewhat secondary reason for establishing that organization. The primary motive was to make a step toward unification.

The IAEA on the other hand was solely aimed at atomic energy. It is clearly stated in the Statute of the Agency that the Agency has two objectives: one, the promotion of the peaceful uses of atomic energy; two, the responsibility for making sure that those peaceful uses are not diverted to military uses. On that basis IAEA was set up in 1956 with headquarters in Vienna. So it has been functioning for approximately ten years.

Let me remind you that the optimism or pessimism about the future of atomic energy and the importance of nuclear power has gone through a number of fluctuations in the last fifteen years. About the time the Atoms for Peace Program started, optimism was very high, and we had statements being made that nuclear power was going to be so cheap that it would not even have to be metered. This turned out to be overoptimistic. We have had nearly ten years between that time and the present, that is, nearly ten years between the time when nuclear power was a technical possibility and the time when it begins to look as if it would be economically feasible in many parts of the world, particularly in this part of the world.

It is very fortunate that we did have that ten years of breathing spell, because international inspection and control is novel and radical. It is difficult to persuade governments that such control is essential. Also, time was needed to develop a system which would be both effective and acceptable.

Actually, the first system that the IAEA developed was in 1960 and was very limited. Even so, it was strongly opposed by the Soviet Union and a number of other members of the Agency. It was only in June of 1963 that the Soviet Union changed its position and became a strong supporter of the IAEA system of safeguards.

Now let me talk about that system in a different way from the way Dr. Seaborg mentioned it. He gave you some of the statistics, and I think the latest figures are a little higher than his in terms of numbers of reactors, numbers of countries, and so on, but that is not important. An important point which has to be made, particularly to an expert audience like this, is that most of the reactors under IAEA safeguards up until the last year or two have been very small research reactors, which may be important in principle, but not in terms of the amounts of material that they might produce. However, as Dr. Seaborg indicated, it is now clear that great production of plutonium is going to occur in this country and in Western Europe in the near future, and that there are enough big power reactors coming along in other parts of the world to produce significant amounts of plutonium. The need for a worldwide system of safeguards is now urgent.

The criteria, as I see them, for international safeguards, are four: (1) efficiency; (2) cost; (3) credibility; (4) acceptability. I want to emphasize particularly the acceptability criterion, because this is different from the domestic situation.

The Atomic Energy Commission can lay down any rules it wants for the control of nuclear materials. There may be grumbling and complaints from the industry, and the rules may have to be modified so that they are not absurdly rigid; they must be reasonably practical and not too expensive. But essentially the AEC is in a position with the help of the Joint Committee to say "This is it; this is what you do."

This is certainly not true in the international area. It is important to remember that we may have to be satisfied with a system of safeguards, international safeguards, that is perhaps 95 or 96 per cent effective in detecting diversion, and is accepted by 98 per cent or 90 per cent of the members of the International Atomic Energy Agency, which is worldwide with about 100 members now. That is much more useful than a system which is 99.44 per cent effective, but which is accepted by only 10 or 20 per cent of the members of the International Agency.

I will tell you a story of our problems that will perhaps illustrate what I have in mind. When the Board of Governors of the IAEA was revising the safeguard system two years ago, we persuaded the group, which had some twenty countries represented, that the inspectors must have access at all times, that inspectors should be able to go to a facility any time they wanted without any previous announcement. This I think is a very essential part of a safeguards system, not only for processing plants, but for reactors.

One of the representatives from a country I will not name said finally he understood this, and this was fine; he accepted the general principle. But

he said, "Of course, in our country for an inspector to get in, he will have to have a visa, and he will have to give two weeks' notice in order to get a visa."

This is the kind of problem that we encounter that I don't think is encountered domestically.

The cost problem is another problem. Even though the Soviet Union supports vigorously the general idea of international safeguards now, they always protest about the costs being borne by the International Agency, as they have been up to the present time. It is not certain that the International Agency can continue to carry all of these costs, but no satisfactory compromise has yet been worked out.

Anything that can be done in the research line that will reduce the cost of safeguards is very important from the international, as well as the domestic, point of view.

Now, credibility is another real problem. It is largely in the interest of credibility that the United States has adopted the policy of transferring the responsibility for administering safeguards from the U.S. to the IAEA in most cases where the U.S. has bilateral arrangements to assist projects for the peaceful use of atomic energy. The great advantage of the IAEA from this point of view is its worldwide membership of nearly 100 countries.

Some of the other problems mentioned in our discussions in IAEA also have a bearing on the question of safeguards research. Some of the countries are worried about interference with efficient operation and about trade secrets. We have done our best in the new IAEA system to guard against these dangers. Many countries also have political problems. They have to be concerned about the nationality of inspectors that may be sent to their countries. They may object to certain nationalities and not want them coming to their countries, but they don't want to object overtly.

We think we have gotten around that by insisting on an informal consultation ahead of time as to what inspectors will be sent from Vienna to Country A or B. But it still is difficult and is something that I don't think is involved in the domestic scene.

I might conclude by repeating Dr. Seaborg's plea for research. It would reduce costs. If we could get methods for solutions of some of the problems which are going to be discussed later on today, which would really work, it would make it much easier to answer the question of interference with operation, stealing trade secrets, and so on. Mind you, I don't think these complaints or questions are really serious. But the point is, they exist. If enough countries are worried about these things, whether rightly

or wrongly, then it reduces acceptability and makes it more difficult to get a worldwide uniform system accepted. Anything that can be done to simplify and automate systems of inspection is very much worth doing.

I might point out that the IAEA itself does have a small research program, but its financial resources are such that it is quite out of the question for it to work on the scale that is necessary to solve some of these problems. I hope that later in this session we will get some suggested answers, or answers to some of the technical problems that would be very useful in promoting the widespread acceptance of international safeguards. As you know, the number of countries accepting such safeguards is gradually increasing. If there should be a nonproliferation treaty which involved the use of the International Atomic Energy Agency, this number would increase greatly. I believe the Agency can be developed to handle such a situation but it will need all of the help it can get to do so adequately. Thank you.

ARMS CONTROL IMPLICATIONS OF SAFEGUARDS

H. Scoville, Jr.
U.S. Arms Control and Disarmament Agency
Washington, D.C.

First, I would like to express the appreciation of the Arms Control and Disarmament Agency to the AEC and the Argonne Laboratory for holding this symposium. I don't think there is any group in the government who is more interested and has more hanging on the safeguards problem than we do in ACDA. Improved research in this area is a critical part of our particular problems, as I will try to outline for you today.

The Soviets have been very much in agreement with the United States on the desirability of achieving a nonproliferation treaty, and a statement to the effect appears, at least from the press reports, to have been at least one of the concrete results of the weekend. There is thus an added push on the part of our two countries to get a nonproliferation treaty tabled at Geneva. (Note: Treaty tabled by U.S. and Soviet Union, Geneva, August 24, 1967.) We have been negotiating this treaty for quite a number of years, but perhaps only last fall did we begin to make real headway. But there has been one difficulty after another in getting general agreement on a treaty which would be satisfactory to everyone for tabling by US-USSR co-chairmen for consideration by the ENDC.

One of the good results of the recent Middle East crisis has been an increased realization on the part of a very large number of people at all levels--not only in this country, but around the world--of the importance of achieving a nonproliferation treaty, and perhaps also a firmer recognition of how important it is that the treaty have a mandatory safeguards article. The safeguards article is needed to provide all parties with confidence that the treaty is being lived up to. While this has been the consistent position of ACDA, the AEC, the Joint Committee on Atomic Energy and others, it has not always been the view of everyone. I think the recent crisis has certainly helped to make the point; none of us would like to go through what we went through during the Middle East crisis if either or both of the participants had nuclear weapons. Even if there were an uncertainty as to whether or not one side or the other had nuclear weapons, the crisis would have been much more difficult to handle. Unless we can get a nonproliferation treaty with safeguards, I think we may well have to face such a situation in the future.

Furthermore, I think it is very important to realize that the nonproliferation treaty may well be the only mechanism by which you can get safeguards in the Middle East. The Israelis already have one reactor, as

you know, which is not under IAEA safeguards. It would seem to be in the interest of the Israeli Government to have such safeguards in order to reassure the world that they indeed are not producing nuclear weapons. However, it would be difficult for Israel to accept IAEA safeguards on this reactor unilaterally, and it has been impossible to arrange bilateral discussions between the UAR and Israel. But the nonproliferation treaty, as a worldwide international mechanism, could draw the Middle Eastern countries into the safeguards system.

Now, as has been pointed out, we have been trying to negotiate such a treaty for quite a while. Probably the thorniest problem in negotiating a nonproliferation treaty has been how to get a mandatory safeguards article into it. There are a number of problems in this. Probably the first and foremost has been that of reducing worry about the intrusiveness and increasing the acceptability of, safeguards. Dr. Smyth has just pointed out the importance of this problem. Several countries are worried that if they have safeguards on their peaceful programs, industrial secrets will be given away. They fear that anything they have accomplished as a result of their research programs will be common knowledge, and that therefore they will lose any competitive advantage they may have.

Actually, we think this is not a realistic concern. Our own nuclear industry has said many times that they would subscribe to safeguards since these are not particularly intrusive or dangerous from the point of view of any industrial secrets. In fact, by the time a reactor is sold abroad most secrets would have to be disclosed. Competitive advantage is not maintained by keeping something secret, but by keeping ahead in the research. Nevertheless, as Dr. Smyth stated, it is not what is real when you are talking about international obligations; it is what people think. We have been pressed on this subject over and over by almost every country we have talked to in connection with the nonproliferation treaty and safeguards. They have stated that they were worried about accepting safeguards because it might disclose their special knowledge. Therefore, the problem of reducing the intrusiveness and making these safeguards more acceptable, is a very critical one in order to achieve a nonproliferation treaty.

I think the second problem which has been most difficult in this area has been that of trying to get agreement on how or what kind of an international safeguards system would be used, and particularly as to how IAEA and EURATOM would interact. It has been the U.S. policy for a long time, and Dr. Seaborg has been a strong exponent of this and stated it publicly many times, that our long-term goal is to have a single worldwide international safeguards system. On the other hand, one is faced with the fact of life that EURATOM does exist; it is a viable organization; it has its own interests, and it has the feeling that if it gave up its safeguarding or if it allowed the IAEA to do safeguarding in its area, then

this might have a serious effect on its overall future. After all, EURATOM has developed over the years a good safeguards system, and technically we feel that it is quite satisfactory and compares favorably with any other. Nevertheless, there is the problem of how you meld this system with that of the IAEA, since the Soviets obviously do not wish to give public support to EURATOM, and are pushing very hard for IAEA as the only organization to carry out safeguards under a nonproliferation treaty.

We have been seeking some formulation which might be acceptable to the Soviets--as well as to all the other countries who will wish to sign the treaty--and yet make use of the system in existence in the EURATOM area. As yet we have not found such a formulation.

The last point that I would like to mention relative to the safeguards aspects of the treaty is that of inequality between the nuclear-weapon countries and the nonnuclear-weapon countries. In the present draft treaty only the nonnuclear-weapon countries would agree to accept safeguards on their peaceful nuclear programs. We have been quite willing to have an article which would place all peaceful programs, whether those of nuclear-weapon or nonnuclear-weapon countries, under safeguards, but the Soviets have held out very firmly against this. They have stated that safeguards for nuclear-weapon countries don't make any sense in a nonproliferation treaty. I must say in the context of the nonproliferation treaty this is probably true, since if you are producing weapons material anyway, it is not really of much technical importance to be able to prove that the material that is produced in your peaceful programs is not being used for weapons. From that limited point of view they have a fairly sound technical point. On the other hand, the Soviet view does not consider the discrimination angle, which in turn ties back again to the problem of whether the safeguarding procedures are really intrusive and a hardship on those countries which are accepting safeguards. If that were the case, then in order to put the peaceful programs of all countries on an equal basis, all countries ought to accept safeguards on their peaceful programs equally.

As I said, we and the British have already put some of our peaceful facilities under safeguards, and we would consider extending this offer to more of our peaceful facilities.

We are also suggesting to the Soviets that they make some move in this direction.

They have already accepted the idea that there would be safeguards on the programs of the Soviet Bloc countries and on any transfers of nuclear materials which the USSR made to nonnuclear countries. For example, if the Soviet Union were to transfer reactor materials to Poland, they are prepared to require that these come under IAEA safeguards. So

they have already made substantial moves in the direction of accepting safeguards, but unfortunately not enough to satisfy completely the nonnuclear-weapon countries.

Now, what is the basic U.S. position on safeguards? First and foremost, we think that we must maintain safeguards which are effective. By effective, I mean sufficiently effective as to have a high probability that any significant diversion would be detected, and most importantly, to deter anybody from carrying out a diversion. However, safeguards procedures are no good if they are not also acceptable. As Dr. Smyth said, it does not do much good to have perfect safeguard procedures as long as only 20 percent of the people accept them. You really have not accomplished much in terms of achieving the goal of nonproliferation. So the intrusiveness of the inspection is a very major factor that needs to be studied in order to make these safeguards acceptable to those countries we wish to have sign a nonproliferation treaty. For this reason, we are very interested in any research which can decrease the intrusiveness by using automation or other techniques, and at the same time maintain the effectiveness which is desired.

I might say that the need for a research program in this area has been recognized and has the support of many foreign countries. As a consequence we now have in the draft treaty which is being considered for tabling a paragraph in the preamble which strongly endorses research programs to devise improved safeguards techniques.

We feel that a large part of the research in this area is going to have to be the U.S. research program. We in ACDA are very much interested in supporting this program, and have been and will continue to work very closely with the Atomic Energy Commission to foster anything that we think is worthwhile in this particular area. We also will try to see that international programs in this area are also pushed forward.

In summary, let me just say that we in the arms control business are extremely interested in what you are doing today. It is a very important part of our program, and an extremely important element in trying to achieve the goal which, I think, we all seek, and that is to prevent the further spread of nuclear weapons. Thank you.

COMMENTS ON THE REPORT OF
THE AD HOC ADVISORY PANEL ON
SAFEGUARDING SPECIAL NUCLEAR MATERIAL

Ralph F. Lumb
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On August 5, 1966, Chairman Seaborg convened in Washington, D. C., an Ad Hoc Advisory Panel on Safeguarding Special Nuclear Material. The Panel was charged to conduct an independent review and appraisal of AEC safeguard policies and procedures for special nuclear materials used domestically and distributed by the United States internationally.

I like to think the importance attached by the Commission to the subject was reflected in the extremely well-balanced constituency of the panel: well-balanced in terms of special talents and extensive backgrounds in the many and diverse subjects which have a bearing on safeguards. It consisted of:

Ralph F. Lumb, Chairman	Director, Western New York Nuclear Research Center, Incorporated Buffalo, New York
Francis P. Cotter	Vice President, Washington Office Westinghouse Electric Corporation Washington, D. C.
Gerald Charnoff	Partner, Shaw, Pittman, Potts, Trowbridge & Madden Washington, D. C.
Paul Grady	Partner (retired), Price Waterhouse & Company New York, New York
Aston J. O'Donnell	Manager of Development Bechtel Corporation San Francisco, California
Louis H. Roddis, Jr.	President, Pennsylvania Electric Company, a Subsidiary of General Public Utilities Corporation Johnstown, Pennsylvania
Fred H. Tingey	Manager, Operations Analysis Idaho Nuclear Corporation Idaho Falls, Idaho

In addition the AEC provided two secretaries, Vincent C. Vespe, Director, Division of Operational Safety, AEC, and Ralph G. Page, Chief, Indemnity and Export Licensing Branch, Division of State & Licensee Relations, AEC, who contributed substantially to effective and efficient sessions of the Panel.

Safeguarding special nuclear materials is an important facet of a major international policy objective of the United States, namely, limiting the proliferation of nations with a nuclear weapons capability. The need for safeguards was well-documented as early as the Baruch plan which was submitted to the United Nations in 1946 and has consistently been considered an essential ingredient in our nonproliferation policy ever since.

Looking ahead a couple of decades, the Panel concluded that the needs for a safeguards program would grow rapidly and the demands upon the program would be severe. The materials and the technology of the rapidly expanding nuclear power programs in many foreign nations can be equally useful in a nuclear weapons program. Further, it seems inevitable that a world market in special nuclear materials will develop, and with such a market one can expect the development of a black market, and attempts by terrorist and insurrectionist groups to turn these materials to their own purposes.

To appraise our ability to cope with these future problems the Panel carried on extensive discussions with representatives of eleven organizations of the Federal Government, two prime contractors of the AEC, four private industries deeply involved in the processing of special nuclear materials, a research institute, a professional society, and two international organizations currently involved in safeguarding special nuclear materials. In addition, visits were made to six sites at which there was extensive processing of special nuclear materials, four representing private industrial corporations and two representative of major AEC prime contractors. The Panel received excellent cooperation and discussions were generally very candid; only once did concern for proprietary information somewhat inhibit the scope of discussion.

The Panel recognized a number of areas which it believed needed modification or reinforcement in order that the United States might be better prepared to meet the needs of the future safeguards program. The basic premise upon which these recommendations were formulated was that safeguarding special nuclear materials was sufficiently important to the best interest of the United States, that all possible techniques and mechanisms should be employed in the program.

By establishing severe criminal penalties and providing rewards for information about diversion of materials for unauthorized use, and the

widespread publication of such penalties and rewards, it is expected that a significant deterrent to criminal activities can be created.

The Panel believed that an increase in stature and an improvement in the coordination of the program could be achieved by consolidating the policy making and overseeing in a single office. The Commission, of course, has already taken steps in this direction, and General Crowson will be the new director of that office effective July 1. Through the formal establishment of a special interagency committee, the Panel hoped to bring to bear routinely upon safeguards problems the information, knowledge, and special talents of those agencies.

The Panel believes that routine access to significant quantities of special nuclear materials is sufficiently important to warrant more than the usual credit check by a bonding agency, and that a check for derogatory information at the national level, such as is represented by L clearance procedures, should be employed.

The Panel noted in its site visits considerable variety in physical protection, and believes that there should be developed common standards mutually acceptable to licensees and the AEC.

Because the physical facilities have such a strong bearing upon the ability to control materials, to take an inventory, and to maintain physical security, the Panel believes that there should be a review of all new construction of facilities to handle significant quantities of special nuclear material, and of all substantial modifications to existing facilities.

An important aspect of the administration of the safeguards program is to have timely information about problems as they evolve; consequently, the Panel recommends that criteria be established for the principal areas of uncertainty in the material control program, such as shipper-receiver differences, materials unaccounted for, quantities discarded, or quantities in inventory in an unmeasurable or unmeasured form. Once appropriate limits have been established, simple mechanisms can be set up to keep the administrative agency informed.

The Panel believes that the future needs of the safeguards program are going to be quite demanding in terms of personnel and funds; consequently, the Panel believes it is going to be essential to optimize the application of resources, and recommends that this be done on the basis of the quantities and the forms of the special nuclear materials handled. For example, from a safeguards point of view, certain quantities are sufficiently small so that they should require no special safeguards provision, whereas others are so large and of such a sensitive nature that they may require full-time surveillance.

The Panel believes that sound safeguards begin at the plant management level and recommended that increased emphasis be given to systems of internal management control within all organizations handling special nuclear materials.

Internationally, the Panel recognizes that the United States is only one member in a community of nations and does not have the ability to bring about desirable changes unilaterally; however, the Panel believes that the U.S. should use its influence to encourage the establishment of an effective universal safeguards system under the International Atomic Energy Agency. To this end it should encourage negotiations between Euratom and IAEA for effective surveillance by the International Agency; encourage acceptance of IAEA surveillance by other countries including the major powers; encourage member nations to make available to the International Agency qualified personnel for reasonable terms, preferably five to seven years; and encourage the international pooling of information regarding diversions of special nuclear materials to unauthorized purposes (actual, attempted, or potential).

While continuing its present policy with regard to Euratom pending an agreement for IAEA surveillance, the AEC should improve its evaluation of the effectiveness on Euratom safeguards program

It seemed obvious to the Panel that the U.S. should increase its research and development activities, seeking to develop techniques which will optimize the applications of its manpower resources in the program; and further, the Panel believed that a well-coordinated research and development program on an international basis could be developed. This symposium is an excellent step toward those objectives.

Because of the severe demands anticipated by the Panel for qualified personnel in the future, and the inefficiencies associated with on-the-job training, the Panel recommends the development of an international school of safeguards which would be utilized by both the International Agency and the national programs in training personnel. Such a school would naturally contribute to research programs, and could soon become a center for the accumulation and distribution of information relating to safeguards.

Because of the high sensitivity of materials, components, and weapons transferred to the Department of Defense, the Committee believes that if there has not been a recent review of applicable safeguards procedures to these materials, it would be timely for such an independent review to be conducted.

These recommendations contemplate the marshalling on an orderly basis, of all available mechanisms, techniques, special talents, and know-how toward implementing an efficient and effective program for safeguarding special nuclear materials.

If appropriately stimulated, I am certain that this audience can make a substantial contribution to this nation's nonproliferation policy. I would not hesitate to compare the challenge of developing new safeguards techniques to placing a man in orbit. If you feel I am exaggerating, just try coming up with a conceptual plan for the tamper-proof remote indication of special nuclear material flows and inventories in a complex processing plant. Such a system would be invaluable to the safeguards program, optimizing use of its resources and minimizing the intrusive nature of the program. It may not have the same aura of glamour as orbiting man, but the challenge is comparable; and we have orbited man.

GENERAL CROWSON: Thank you. Our next speaker is Dr. Ted Taylor, who is a consultant to the Division of International Affairs of the Atomic Energy Commission. His topic is "Scientific and Technical Needs for Safeguards."

Just a word about Dr. Taylor. He has been on both sides of the fence, and by that I mean he has been in the weapons program as well as the safeguards program, so I hope he gives a balanced presentation.

TECHNICAL REQUIREMENTS FOR NUCLEAR SAFEGUARDS

Theodore B. Taylor
Consultant to the U.S. Atomic Energy Commission
Washington, D.C.

The most important requirement for nuclear safeguards research and development is a clear definition of what nuclear safeguards are meant to do. I have been tempted to plan to make that statement and then sit down, but that isn't very constructive. So I shall try to tell you why I think this definition is needed, suggest some specific possible nuclear threats that should be taken into account in the definition, and briefly discuss a few things that I think ought to be done.

Let me use part of the IAEA Statute to illustrate some of the difficulty. It states that its safeguards are "designed to ensure that special fissionable and other materials, services, equipment, facilities, and information made available by the Agency or at its request or under its supervision or control are not used in such a way as to further any military purpose." One might use that, as a start, for a definition of safeguards. But immediately there are difficulties. The word "ensure" strongly implies, among other things, protection from theft. And yet people in the IAEA, Euratom, and the AEC disagree about the extent to which these agencies should provide or demand adequate physical security for special nuclear materials, especially those they do not own. "Ensure" also implies to me that the Agency should try to recover any lost plutonium, or at least try to find out where it is. But most people believe that the Agency safeguarding responsibility ends after detecting a loss and reporting it to the Board of Governors. I am strongly convinced that tritium, which one might include under "other nuclear materials," should be specifically under safeguards. Others strongly disagree. The word "any" in the phrase "further any military purpose" implies a degree of restriction that no one I have talked to says should be taken literally. How does one define a military purpose? I suggest that nuclear safeguards would be no more significant than safeguards of any other valuable materials if uranium or plutonium could not be used for making very powerful explosives or radiological warfare agents. I believe it would be better not to mix the IAEA's dedication to the development of peaceful uses of nuclear energy with the specific definitions of the purposes of nuclear safeguards.

Even greater difficulties appear when one tries to be quantitative about safeguards--to decide what surveillance accuracies to aim for, for example, or how much special nuclear material is an unreasonably large stockpile for a facility or for a country, or how far to go in insisting on physical protection of materials from theft. The Agency Statute uses the phrase "so far as it is able" at one point, but that does not help in planning for the future. The cumulative amount of plutonium produced by all the

world's power reactors will exceed 100,000 kg in about five years, and the rate of production by the end of 1972 will exceed 30,000 kg/yr. Just one percent of either of these numbers potentially represents many bombs and a lot of money. Does this mean that surveillance accuracies must be very much better than one percent? Before one says obviously not, I suggest he think carefully about what the dangers are, and how they may be affected by the accuracy of the surveillance system.

Suppose that some safeguards system can detect, with 90% probability, the diversion of 10% or more of the special nuclear materials in a country, and, say, with 10% probability, 1% or more of the material. Such a system could, I believe, ensure that a non-nuclear power would not secretly build up a completed military nuclear stockpile. But it would not prevent or even necessarily strongly inhibit the undetected diversion of material for a few bombs by some non-national group of people. Neither would the system prevent stockpiling of large quantities of special nuclear materials that could quickly be incorporated into a prefabricated stockpile of nuclear explosive devices that have everything except the required special nuclear materials. Nor would the system, by itself, prevent the theft of large quantities of plutonium, uranium, or tritium by massive armed robbery.

All that is required to make a crude fission explosive that can be carried in an automobile is a few kilograms of plutonium or highly enriched uranium, a practical scientist or engineer who is familiar with the basic, unclassified properties of chain-reacting systems, and some other components that could easily be made and assembled in any city in the world. Devices of this kind, in the hands of unscrupulous revolutionary or criminal groups, could be used for severe threats against national governments or other very large groups of people, in forms for which a strategic deterrent force is useless. An unusually large assembly of national government leaders in the United States Capitol or the Kremlin, for example, could be killed by the explosion of such a device in a parking lot near the building. An organization that does not have to identify itself could try to force a government to take certain action, using the threat of detonating covertly assembled nuclear explosives in several of the nation's big cities, if the demanded action were not taken. The United States could be asked to pull military forces out of Viet Nam, for example. There are several quite different organizations that would like to see that happen.

One can think of many other examples of threats, using not only relatively low-yield, crude fission explosives, but also much larger-yield thermonuclear devices, that could make use of diverted or stolen special nuclear materials in the hands of groups or people that need not either represent any nation nor identify themselves. Furthermore, plutonium, highly enriched uranium, and tritium are all extremely valuable materials, worth much more than gold per unit weight, because of both their commercial and their military value. These materials are bound to become

temptations to unscrupulous criminal organizations that are now sustained by international illegal markets in gold, diamonds, dope, or arms. These are some of the other dangers, much less publicized than the dangers of further proliferation of national military nuclear stockpiles, in the development of large-scale industrial use of nuclear energy without an effective system of international nuclear safeguards.

I have discussed these matters that may seem out of place in a talk about the technical requirements for safeguards because I believe they have to be thought about very hard before we can be reasonably sure that we know what an effective system of safeguards should do. But even after all the important classes of dangers in an inadequately safeguarded nuclear energy system have been thought through, there are still some other constraints, often nontechnical, that should be taken into account in establishing a set of detailed requirements for a safeguards system. I haven't time here to do much more than mention some of these.

One constraint is the concern of many people that safeguards will interfere with the development of the nuclear power industry. Fortunately most, if not all of the information required for effective safeguards is also useful in the development and operation of the nuclear-energy industry. Some types of required physical security measures may, in some sense, interfere with the industry; in those cases I certainly believe that industry should yield, not the security measures.

Another constraint is cost. I strongly disagree with the idea that low costs for nuclear power should come first, effective safeguards, second. But, fortunately, it looks as though effective safeguards can be achieved at costs that would add one percent or less to the costs of nuclear power, and there is a good chance that even these costs would pay for themselves through the benefits that many safeguards measures hold for the industry.

A third constraint is the availability of adequately trained people as inspectors, especially within three or four years, when very large numbers of inspectors are likely to be required. This constraint tends to place emphasis on reliable, automatic data recording, transmittal, storage, and analysis techniques and on safeguards procedures that can be standardized.

Related to this is a fourth constraint that the safeguarding process has to be inspiring enough to make possible recruitment of good technical people to oversee inspections and research and development work.

A fifth constraint has to do with industrial secrecy. Of two otherwise equally effective safeguards measures, the one that requires the least detail in the description of components or processes used in the nuclear-energy industry is the more acceptable.

Finally, there are different types of political constraints, some of which are closely connected with ones I've mentioned, and many of which are extremely hard to define. Political constraints, if they are strong and persistent, usually have very important effects on the technical requirements for safeguards. A history of recent political instability in a country, for example, has a rather direct bearing on the quantities and physical forms of special nuclear materials that should be allowed to accumulate in that country. But other political considerations may make it impractical to take such a connection into account.

Having talked about dangers and difficulties and constraints, I want now to try to be more constructive and sketch very briefly a few concepts that I think would be useful and perhaps even necessary parts of the planning or operation of any effective safeguards system. I want to emphasize international nuclear safeguards, but I think that most of what I have to say also applies to national safeguards. This is by no means an attempt to summarize all the basic requirements for specific safeguards systems.

First, we need to develop and then use an information collection, storage, manipulation, and retrieval system that can keep track of all the important parameters that define the past, present, and future state of the world's nuclear-energy system. I do not believe that anyone now knows or can easily find out enough about the way the world's nuclear-energy system is developing to have a clear idea of what it is that we will want to safeguard in, say, three or four years. You all know how extremely fast things are now changing. This information system should make it possible to determine past, present, and future rates of production and consumption of special nuclear materials classified according to specific facilities, types of facilities, manufacturers, nuclear-energy complexes, countries, physical and chemical form of the material, etc.

Second, we need to develop suitable techniques for describing an international nuclear safeguards system on paper or in electronic data-storage units. These should provide general-purpose formats and programs that would be suitable for a wide variety of types of systems.

Third, we need systems-analysis techniques that are suitable for studies of the effectiveness of specific safeguards-system concepts. I believe the techniques should be suitable for deriving at least the following:

- a. The accuracy with which special nuclear materials are balanced, classified according to the type of material, the history of the material between inventories, geographical regions, types of facilities (e.g., fuel-element fabrication, transport, power plants, reprocessing plants), countries, international organizations, etc. The capability to make parameter sensitivity studies is essential.
- b. Operating costs of the system.
- c. Capital costs of the system.
- d. Numbers and types of safeguards personnel required.
- e. Effects of the safeguards system on the nuclear-energy system, in terms of cost increases or decreases, production delays, increases or decreases in safety parameters, for example.
- f. Levels at which diversion could take place without detection, organized by type of facility, energy system modules (combinations of fuel-element production, power plants, and chemical reprocessing plants, for example), industrial organizations, countries, international organizations, type of material and their physical and chemical form, etc. This implies that the analysis techniques include the ability to play diversion games on a machine.
- g. Changes in elements (components or procedures) of the nuclear-energy system that would increase the effectiveness of safeguards. The technique should also be suitable for deriving the effects of such changes (changes in costs, delays, or even improvements) on the energy-production system.
- h. Changes in parts of the safeguards system that would increase its effectiveness.
- i. Requirements for further safeguards research and development.
- j. Requirements for further R&D on elements of the nuclear-energy system to make it easier to safeguard effectively.

Fourth, an electronic information storage and retrieval system for use by the safeguards organization in its operations is needed. In summary, this should be capable of telling the safeguards personnel where and in what form all the special nuclear materials for which they are responsible were, are, and will be, as rapidly as is possible, by reasonable use of modern data acquisition, transfer, storage, processing, and display techniques.

Fifth, uniform reporting procedures and a hardware system that transmits data rapidly, in the right form, to the safeguards data bank are both required. In practice, provision should be made for reports from inspectors, facility operators, and automatic surveillance-recording equipment. The system will probably require several different types of data transmission links.

Sixth, there is a need for computing-machine programs for analysis of the implications of deviations from expected values of data reported through the safeguards information storage and retrieval system, and to test the data for internal consistency (comparisons between measurements and calculations, for example). Many of the sampling techniques will be based on statistical methods that are very complex, but that can greatly increase the effective accuracy of a surveillance system if the data are completely analyzed.

Finally, having so far talked mostly about information-system software, I want to give just one example of a specific class of measuring systems that I think looks very attractive, but which will require much theoretical and experimental work to develop. I call this a fuel-element isotopic-content scanning and tracking system. The heart of the system is any one of at least three possible techniques for nondestructively measurement of the relative and absolute amounts of fissionable isotopes in new or in irradiated reactor fuel elements. The techniques I have in mind use a zero-power critical assembly, a pulsed neutron source, or an electron linear accelerator. The first technique, which was examined in considerable detail at Westinghouse four or five years ago and, I believe, still looks promising, is based on measurements of the reactivity changes of portions of the fuel element as it is inserted into an annular-core critical assembly. The second, which Bob Keepin from Los Alamos will describe shortly, is based on observation of the delayed neutrons from fission products generated by an intense pulse of neutrons that strike the element. The third technique, which Bob Beyster from General Atomic will describe later today, is based on the observation of gamma rays or neutrons from fission of fuel-element isotopes by a short, intense pulse of gamma rays generated by impact of a pulse of electrons on the fuel element. All three techniques show some promise of working even in the gamma-ray background of irradiated fuel elements. All the techniques are suitable for use in a way that very accurately compares measurements of the properties of unknown elements with known standard elements, measurements of the same element before and after irradiation, and fuel-element measurements and prediction calculations.

One can imagine that the fuel-element scanning devices are permanently installed and used routinely near the point of output of elements from fuel-element-fabrication plants and near the point of input to chemical reprocessing plants. Each fuel element, defined as the smallest assembly into which fuel is separated between packaging for shipment from the

fabrication plant and destruction at the reprocessing plant, is somehow identified, kept under surveillance--either actively or by a system of seals and physical constraints--at all times during its life. Its whereabouts and detailed irradiation history are recorded in the safeguards-system data bank. Finally, a four-way comparison is made between measurements before irradiation, measurements after irradiation, measurements of a known standard set of fuel elements, and calculations that use the irradiation-history data on both the fuel-element positions in the reactor core and the reactor power level. By use of such techniques there is at least a chance of achieving very high precision in the overall material balance accounts between the input and output of fuel-fabrication plants and of chemical reprocessing plants, as well as preventing essentially any undetected diversion of material between the output of the fabrication plant and the input to the reprocessing plant, since the latter requires only counting and watching or constraining a finite number of specific items.

Just one comment about cost. The operating costs of the types of scanning facilities I have been suggesting could run as high as several hundred thousand dollars per year, and still represent 1% or less of the value of the annual throughput of U^{235} or plutonium in reasonably large plants. The write-off of capital costs, at first glance, looks comparable.

Careful study of this kind of a safeguards concept, if one accepts that it would be useful, will yield a large number of rather specific requirements for applied research into sources, measuring techniques, seals, shipment and packaging procedures, chemical analysis techniques, development of theoretical calculation techniques, and a lot of basic physics. In view of the time, I leave this as an exercise for the listener!

Many of you may think I have painted much too grandiose a picture of what an international nuclear safeguards system may become, that we cannot afford the costs and manpower to develop and operate them. To those I simply want to say two things. One is that the explosion in the middle of New York City of one large, dirty thermonuclear device, perhaps secretly assembled there, would kill several million people by fallout, would destroy many tens of billions of dollars of property, and would be very likely to start World War III. The other is that the nuclear-energy system is now a multibillion dollar a year enterprise, and increasing rapidly.

ORGANIZATIONAL CHANGES WITHIN THE AEC AFFECTING SAFEGUARDS

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I think first of all it might be well to describe what the current organization is, and then show the differences in the new proposed organization to take effect July 1, 1967. The organizations currently responsible for performing the safeguards function are listed below with a summary of the functions performed (see Fig. 1).

Fig. 1. Organization Prior to July 1, 1967.

- a. Division of Nuclear Materials Management (DNMM) and Field Office Nuclear Materials Management Staffs

Functions:

- (1) Develops a materials-quantity data system for timely reporting to the AEC of all nuclear-material transactions, inventories, consumption, and losses. For that material owned by the AEC, provides the quantitative data for the planning,

programming, budgeting, costing activities, and financial controls, such as billing for losses and consumption, and developing inventory values.

(2) Develops policies, standards, and procedures; audits nuclear-material records; verifies inventories; analyzes losses and loss mechanisms; evaluates technical validity of measurement methods of:

(a) AEC license-exempt facilities for both safeguards and contractual purposes;

(b) AEC-licensed facilities, effective August 1, 1966, for safeguards purposes to ensure conformance to regulatory requirements;

(c) AEC-licensed facilities for contractual purposes.

(3) Administers programs for the development of chemical and isotopic standards and for research and development, leading to improved statistical and measurement methods.

(4) Coordinates the development of short-term forecasts of special nuclear-material requirements for peaceful and military reactors and research and development to plan operation of the AEC's production facilities.

(5) Develops the distribution control system and policies to optimize the use of special nuclear materials.

b. Director of Regulation

Functions: The Director of Regulation is responsible for administering safeguards of material in the possession of licensees.

c. Division of International Affairs

Functions:

(1) Responsible for safeguards functions applicable to materials distributed abroad to foreign governments or other international organizations for nonmilitary purposes.

(2) Development of safeguards policies and arrangements with international organizations expected to participate in safeguards controls.

d. Division of Security

Functions: Develops policies, procedures, criteria, and standards for protecting facilities, equipment, and materials of security interest or strategic importance to the AEC in the possession of contractors (both licensed and unlicensed) who under the terms of their contracts are required to protect such material and are not held financially responsible for the loss of such material. Performs related inspections and appraisals.

The Assistant General Manager for Administration has responsibility for two groups involved in the safeguards program, the Division of Nuclear Materials Management, headed up at this time by Douglas George, and the Division of Security.

The Division of Nuclear Materials Management is responsible for developing policy and procedures in the area of materials management or safeguards for both the nonlicensed contractors and for licensed AEC contractors. Until this past year, the Commission had not imposed requirements on licensees through the Code of Regulations, as to the kind of control that should be exercised over their nuclear materials.

Also reporting to the General Manager is the Assistant General Manager for International Affairs, and under him the Division of International Affairs, which is responsible for the development of policies associated with our international safeguards system. He is also responsible for conducting inspections required under our international program, that is, the inspection in countries under bilateral agreements with the United States Government.

Also reporting to the General Manager is the Special Assistant for Disarmament, who also is an advisor to the Commission, and who maintains liaison with the Arms Control and Disarmament Agency.

The Director of Regulation has had responsibility for licensees, but as I pointed out, until this year there have been only minimal requirements imposed upon our licensees, and those were largely through leases and contracts.

Field offices are responsible for the inspection of nonlicensed operating contractors of the Commission, and have also carried out the inspections made at AEC-licensed contractors and of other licensees under regulations that have been developed in the past year.

That briefly is the current organization.

The new distribution of Safeguards functions, which will become effective July 1, 1967, is as follows (see Fig. 2):

Fig. 2. New Organization for AEC Safeguards as of July 1, 1967.

I. Office of Safeguards and Materials Management

A. The Director, Office of Safeguards and Materials Management, reports to the General Manager, but provides advice and counsel directly to the Commission on safeguards matters (both domestically and internationally). Specifically the Director:

1. directs the activities of the Office;
2. provides periodic reports to the General Manager, the Director of Regulation, and the Commission on the status of the safeguards program;
3. recommends to the General Manager, the Director of Regulation, and the Commission:
 - a. corrective action in connection with activities of licensees, nonlicensed contractors, and parties to international agreements; and
 - b. suspension of such activities and/or arrangements when necessary;

4. chairs the Advisory Committee on Nuclear Materials Safeguards.

B. The Office of Safeguards and Materials Management:

1. develops AEC-wide policies, procedures, and standards for the safeguarding of special nuclear material, domestically and internationally,* in coordination with the Director of Regulation, Division of International Affairs, Special Assistant for Disarmament, Division of Security, other AEC components, and other Government agencies;
2. issues directives applicable to field offices and nonlicensed contractors;
3. identifies need for and conducts research and development in support of AEC safeguards program;
4. conducts periodic review and appraisal of the AEC safeguards program, including
 - (a) review of AEC staff survey reports and activities, and
 - (b) spot checks of inspections of nonlicensed contractors and licensees as required;
5. conducts special investigations of activities of licensees, nonlicensed contractors, and parties to international agreements as required;
6. in connection with activities of licensees, nonlicensed contractors, and parties to international agreements,
 - (a) recommends corrective action to assure compliance with overall safeguards policies, procedures, and standards, and
 - (b) recommends suspension of such activities and/or arrangements when necessary;
7. provides staff support for the Advisory Committee on Nuclear Materials Safeguards;
8. provides data for reports to be made to the General Manager, the Director of Regulation, and the Commission on the status of the safeguards program;

*Does not include material transferred to DOD under Section 91(b) of the Atomic Energy Act of 1954, as amended.

9. provides staff guidance and assistance with respect to policies, procedures, and standards developed by the Office;
10. conducts on-site safeguards inspections of material distributed abroad under bilateral agreements;
11. compiles data for short-term forecasts of requirements for AEC and licensees' peaceful and military research and development programs;
12. maintains records and makes necessary reports regarding source and special nuclear materials, including materials distributed abroad.

II. Division of Nuclear Materials Safeguards

A. The Director, Division of Nuclear Materials Safeguards, reports to the Director of Regulation and is responsible for administering safeguards for all material held by licensees regardless of its origin (including Government-owned material in licensee plants).

B. The Division of Nuclear Materials Safeguards:

1. participates in development of policies, procedures, and standards for safeguarding nuclear material;
2. develops regulations based on policies, procedures, and standards furnished by the General Manager;
3. reviews and approves licensee safeguards program and incorporates essential elements as condition of license;
- 4.* conducts surveys, audits, and inspections of licensee facilities for compliance with safeguards requirements;
- 5.* conducts special investigations of licensee facilities where "material unaccounted for" (MUF) at unaccepted level or diversion suspected;
- 6.* takes enforcement action, including suspension of license where warranted;
7. provides the Office of Safeguards and Materials Management copies of survey reports and other reports of safeguards activities as required to keep the Office fully informed of the status of the safeguards program.

*May be assigned to Division of Compliance at appropriate time.

III. Special Assistant for Disarmament

1. participates in development of safeguards policies;
2. advises on all matters involving arms control and disarmament as they relate to safeguards;
3. provides the Office of Safeguards and Materials Management with reports of safeguards activities as required to keep the Office fully informed of the status of safeguards program.

IV. Division of International Affairs

1. participates in the development of safeguards policies applicable to material distributed abroad;
2. negotiates and maintains liaison with Department of State, foreign governments, and international organizations on safeguards policies developed by the Office of Safeguards and Materials Management;
3. provides the Office of Safeguards and Materials Management with reports of safeguards activities as required to keep the Office fully informed of the status of the safeguards program.

V. Division of Security

1. participates in development of the aspects of safeguards policies dealing with physical protection of materials;
2. provides functional direction and reviews field administration of physical security policies applicable to nonlicensed contractors;
3. provides the Office of Safeguards and Materials Management with reports of safeguards activities as required to keep the Office fully informed of the status of the safeguards program.

VI. Field Offices

1. implement AEC safeguards and materials-management policies, procedures, and standards applicable to nonlicensed contractors, including
 - (a) reviewing and approving of safeguards and materials-management programs;
 - (b) conducting surveys of the nonlicensed contractor's facility;
 - (c) taking corrective action where appropriate;

2. provide the Office of Safeguards and Materials Management copies of survey reports and other reports of safeguards activities as required to keep the Office fully informed of the status of the safeguards program;
3. carry out necessary contract administration associated with special nuclear material distributed to licensees.

The new organization, as has been described to you by Dr. Seaborg, creates a new Office of Safeguards and Materials Management, and the Director of this office, General Crowson, will wear two hats. He will report to the Commission as an individual, advising the Commission on safeguards matters, and he will also report to the General Manager. In this latter capacity, his office will be responsible for developing all safeguard policies, procedures, standards, and criteria across the board, both in the international and domestic areas.

The Office of Safeguards will also appraise the performance of those who will be responsible for implementing the policies and procedures developed by this office.

General Crowson will be responsible for the conduct of whatever research and development is sponsored by the AEC in both the domestic and international areas.

The Safeguards Office will conduct certain inspections now that have been the responsibility of DIA.

Finally, the Safeguards Office will be in the position of reporting to the General Manager and to the Commission of any corrective action it sees fit to take with respect to both nonlicensed contractors and licensees.

I will continue to have the Division of Security. It will continue to perform a service to the Office of Safeguards in connection with development of physical security standards to be applied both to nonlicensed contractors and licensees. The Division of Nuclear Materials Management will be absorbed into this new Office of Safeguards headed up by General Crowson.

The Division of International Affairs currently has 13 positions which will be reduced to about four. They will continue to have the responsibility for negotiating with international organizations in foreign countries, using the policies, procedures, and standards developed by the Office of Safeguards. They will continue to maintain liaison with the Department of State and with the other international organizations, such as Euratom and the IAEA.

Field offices will continue to carry out the inspections of nonlicensed contractors. They will implement the policies developed by this Office with respect to cost-type nonlicensed contractors and perform the necessary inspections.

The Special Assistant for Disarmament will continue to function as he has been functioning under the current organization.

The other significant change is under the Director of Regulation. A new Division of Nuclear Materials Safeguards will be established, and this division will incorporate the policies, procedures, and standards and criteria developed by the Safeguards Office into regulations, and then conduct inspections of licensees.

I failed to mention earlier that we now have two committees advising the Commission (the Divisions of Nuclear Materials Management and International Affairs) on safeguards and nuclear-materials management. We are proposing to abolish those two Committees and create one new one that will report directly to the Commission.

DISCUSSION

DR. KEEPIN: It appears that General Crowson appears in two places--Director of the Office of Safeguards, and Chairman of the Advisory Committee.

MR. VINCIGUERRA: He is an individual advising the Commission, and he is head of the office reporting to the General Manager.

DR. KEEPIN: Has the Director of the Division of Nuclear Safeguards been named?

MR. VINCIGUERRA: Not as of this time.

DR. KEEPIN: The Advisory Committee has not been named, except the Chairman?

MR. VINCIGUERRA: This committee is in the process of being named at the present time.

DR. KEEPIN: Who is the Assistant General Manager for International Affairs?

MR. VINCIGUERRA: Myron Kratzer. He wears two hats also.

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NUCLEAR MATERIALS CONTROL SYSTEMS STUDIES

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Introduction

This paper devotes primary attention to national systems of control of nuclear materials rather than individual measurements or analyses of segments, e.g., a power reactor or a fuel-fabrication facility. A proper system of materials control is a collection of observations of the quantity and quality of nuclear materials, organized to provide a unified and concise summary of the location and amount of materials in the system. The system's ability to show discrepancies between the quantity and quality of materials assumed to be on hand and that actually found is important to materials safeguards. The system does not prevent diversion--it does present an early warning of possible diversion to the materials manager. The system validity is determined basically by the quality of the data from which it is constructed, but the system itself varies in usefulness. The system utility is determined by its timeliness, precision, accuracy, detail, and the mode of presentation of the collected data.

In this paper, a summary of the current practice and future requirements of the U.S. nuclear-materials control system is followed by a brief review of the more important systems research reported to date. Finally, some recommendations for future research are made.

Background

Nuclear materials are a unique class of the many materials subject to inventory control. They are unique because they contribute to, or are essential parts of, nuclear weapons and, although many of them have very high unit costs, they are produced in large quantities. The relationship between cost and quantity of common and nuclear materials is illustrated by considering an annual U.S. production of 58 short tons of gold at slightly over \$1 million per ton, 1150 tons of silver at \$26,300 per ton, approximately 1000 tons of zirconium sponge metal at \$12,000 per ton, 7000 tons of titanium sponge at \$2,600 per ton, and 500 million tons of coal at an f.o.b. mine value of about \$4.50 per ton, and comparing this with about 10,000 tons of uranium concentrate at \$12,000 per ton* and a U.S. capacity for producing about 80 tons of enriched (90%) uranium at about \$10 million per ton.**

*Tonnages and prices were abstracted from Minerals Yearbook, Vols. I and II, 1965, U.S. Department of Mines, U.S. Government Printing Office, 1966.

**Computed from data given in U.S. AEC press release, K-145, June 14, 1967.

The control systems established for nuclear materials also have unique aspects. While commercial materials are controlled only because of their monetary value, nuclear materials are controlled because of their importance to the national defense and the strategic-political balance between nations. Inventory control is imposed on non-nuclear materials at a single plant, or, at most, the control spans the plants of a single commercial organization or military service. Nuclear materials are frequently subject to control or control review by national and international bodies. Furthermore, while control systems for non-nuclear materials consider chemical and mechanical processing with attendant scrap and wastage losses, national nuclear-materials control systems must consider nuclear transformation, isotopic enhancement, and radiation decay, as well as many chemical and mechanical steps.

The existing U.S. nuclear-materials control system has grown since the Manhattan project of 25 years ago, when it consisted of individual plant-control activities. The existing system has drawn heavily on a long tradition of materials monitoring and control applied to coal, steel, gold, wheat, munitions, automotive parts, and many other items. The system was directed first to the control of secret or "strategically valuable" items; the effort and attention devoted to control were not directly related to cost of production or value as are most industrial efforts of materials control. In most past and present operations, the material is government-owned; it is processed in government-owned plants; and the plant operator, while accountable for its proper control--or loss if this occurred--is not financially responsible. In these operations, contractor management is responsible for setting up the individual control systems. The individual systems and their operations are reviewed by the AEC operations offices. These offices accumulate summary data on inventory and transactions, which are further summarized and reviewed by AEC headquarters. Materials processing, production, and movement are controlled by the government, and processing and production operations are conducted at few sites.

Some modifications have been made to accommodate materials leasing and the government-owned materials processing in private facilities, with the private operators accepting financial responsibility. In the past, if material was lost, the cost to the operator was assumed to be a dominant control incentive. The Lumb panel has recognized the necessity of additional control when national interests dictate.¹ The Commission also has recognized this in recently published regulations. These regulations are necessary to safeguard national interests under conditions of private ownership.

Materials-control systems of varying complexity and completeness have been constructed to serve the needs of Great Britain, France, presumably the USSR and China, as well as the Western European Union, the European Nuclear Energy Agency, Euratom, and the International Atomic Energy Agency.²

More changes--some of them drastic--are expected in the future: first, a growing civilian nuclear power economy will place larger quantities of materials in many more locations. A capacity of 150,000 MWe in 1980 also will produce many more transactions than are encountered at present. In addition to an increase in quantities of data and numbers of sites and reports, the future materials-control system must deal with new manufacturing and processing techniques, which might include gas centrifuge enrichment, automated fuel fabrication, breeder reactors with attendant differences in fuel characteristics, and volatility reprocessing. For all of these systems, the materials controller will have few precedents to guide him. For example, the plant operators will not have the benefit of past experience in judging normal operating loss or in evaluating book-physical inventory discrepancies in new reprocessing plants. The operating characteristics of the reactors will be sufficiently different that the reactor physicist may have bias in his calculation of total plutonium produced and its isotopic content.

Second, because the civilian material will soon be privately owned, the inventories held, quantities processed, and shipment rates will be set largely by the economic considerations--sometimes conflicting--of individual corporate managements rather than by the government.

Third, while the AEC, its contractors, and private industry have been creating a revolution in atomic energy, others have been creating a revolution in data handling and data systems. Any changes made in the current system or any new system will surely consider this fact.

Past Research

In contrast to the extensive physical research activities,³ little research has been done on systems development. Some past research has defined the needs of materials and arms-control systems in general terms. Such studies often have considered control in an international environment--a situation that is more complex than the national control problem because it must consider the possibility of large-scale diversion as well as common defalcation. (The national and international control schemes are not essentially different.) Included in this category are studies by Wright,⁴ Beaton,⁵ and the staff of the Johns Hopkins School of Advanced International Studies.⁶ Such studies discuss number of inspectors, etc., without specifying what is controlled or how well. More specific studies that consider the materials-control needs of specific plants or operations have been conducted by the staffs of the Phillips Petroleum Company and the General Electric Company, who discussed nuclear materials control in research and test reactors,⁷ power reactors,⁸ and fuel-reprocessing plants,⁹ and by de la Garza and Milone,¹⁰ who discussed diffusion-plant materials control. Another semi-quantitative study of the Hanford complex by Bennett *et al.*,¹¹ related requirements for inspection and control to the degree of control assurance that might be achieved.

Bennett considered cases of complete and partial duplication of measurements and analysis. The primary quantitative factors were based on Hanford experience and on propagation-of-error techniques. Estimates of additional surety afforded by the presence of the inspection team and its ability to observe plant operations also are given. A more recent study of materials control in fuel-reprocessing plants was made by Granquist and Schneider.¹² Plant design and operations were explicitly considered in relation to safeguards. The accuracy and precision of typical plant measurements are given, as are data on Hanford and Idaho Chemical Processing Plant materials balance and measurement variations. Other studies relating to fuel fabrication and reprocessing facilities are in progress.³

Kinderman and co-workers have outlined materials-control systems, but have not made explicit studies of the control achievable by imposing various techniques. They have suggested that control be considered on a systemwide basis for (1) the allocation of limited and valuable inspection and control resources,^{13,14} (2) the preparation and consolidation of transactions, inventory, and other reports,¹⁵ and (3) the analysis of potential materials loss and the distinction between the discrepancies caused by measurement uncertainties and actual loss.¹⁵ Coggshall¹⁶ has outlined, and currently is seeking to develop, techniques that improve the precision of statements concerning the total amount of materials present in the control system. He first devises alternative statistical models for determining the precision, each involving slightly different sets of measurement data and methods of computation. He then applies mathematical programming techniques to find the allocation of a limited measurement expenditure that gives the best overall precision.

Future Research

What more needs to be done in the development of materials-control systems? Several research projects are suggested that should be attempted in the near future. One is the further development of a centralized data bank where all transactions are speedily recorded and the transactions used to establish timely book inventories at all locations in the controlled system. If possible, the data bank should be compatible with the data systems of all major special nuclear facilities. This centralized system should compare all shipment and receipt data, and test them for significant differences existing between shipper and receiver. The system also should test for bias by comparing receipts at various locations for a single shipper and shipments from a single location to various receivers. The mathematics for this has been outlined by Dresch,¹⁵ but it must be expanded and extended--a second research task. Inventory data accumulated by the system can be used to test reported inventories. The probabilistic models used to check shipper-receiver data also can be used to check the statistical reasonableness of book-physical inventory differences and data on unaccounted for materials. Again, the techniques must be tested, expanded, and applied to a computer format.

The losses from operations that might occur without detection and the rates of diversion below which detection is unlikely have been evaluated by de la Garza¹⁰ and Bennett¹¹ for important parts of a national control system. Drukey¹⁷ also considered losses and alarm levels for an international control system applied to research and power reactors and some supporting facilities, but no one has constructed a model of, and evaluated the losses possible in, a complete national or international system.

With such a model, one could:

1. Establish the possible losses from the entire system under various operating conditions.
2. Test subsystems such as a fuel-fabrication or fuel-reprocessing plant.
3. Take a semiquantitative check on our currently intuitive judgments that have selected certain inventories, transactions, and measurements as being most important. With such data, one can allocate properly the effort placed on the various elements of the control system. For example, the analysis might indicate a potential loss in fuel-fabrication facilities; among the corrective measures would be improvement of accuracy and precision of measurements, duplicate independent measurements, or the presence of inspectors. The effectiveness of the first two of these measures could be tested by model simulation or mathematical analysis.
4. Demonstrate forcefully the necessity for inventory reduction and/or conversion of inventory to accurately measured form.

Development of the model is a third important task in the overall research necessary to develop proper materials control systems.

The suggested tasks are difficult ones. If they are to be accomplished before the present nuclear-materials-control system and its managers are overwhelmed by the detail arising from the rapid growth of domestic civil power, they must be tackled by enthusiastic and inventive people. More than one research contract for each task might well be appropriate, because the timely development of effective systems will be useful in the wider context of international affairs, where the potential proliferations of national civil power and nuclear weapons manufacture are closely linked. Surely credible nuclear-materials-control systems with demonstrated ability to assess potential diversion or accidental loss in complex industrial and national undertakings quantitatively can be adapted to effective international arms control.

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DR. McGONNAGLE: The next speaker on our program this morning is Carl Bennett, who manages the mathematics division at the Pacific Northwest Laboratory of the Battelle Memorial Institute.

| SAFEGUARDS SYSTEMS STUDIES*

21:30600
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(Paper presented by Mr. Bennett)

Background

The Pacific Northwest Laboratory of Battelle Memorial Institute, formerly the Hanford Laboratory, has, since late 1956, been involved in studies aimed at determining the feasibility of and evaluating the effectiveness of inspection and control systems for monitoring the production of nuclear materials. Our first involvement with this subject had to do with disarmament, and, in particular, with the problems that would arise if military production were curtailed and if the operating plants were limited to producing nuclear material for peaceful uses only. During the first six months of 1957 a complete study was prepared of the inspection and control of an operating plutonium complex, including fuel-fabricating facilities, reactors, and separation plants. Similar studies of a gaseous-diffusion complex were prepared at Oak Ridge. Several levels of inspection were considered. Since this report was based on Hanford technology, it was classified "Secret." In late 1959 a declassified version of this report¹ was published, which was subsequently used in disarmament discussions with Great Britain. This report has formed the basis for much of our later work. Following the conversations with the British in 1959 and the work of an interagency task force in March of 1960 in preparation for a presentation at Geneva, reports were prepared on other aspects of the control and inspection problem. This phase of activity culminated with our participation in an advisory and working capacity in the deliberations of the Perkins Panel in the spring of 1961.

Following the formation of the Arms Control and Disarmament Agency, two further reports were produced on behalf of the Office of Disarmament of the Atomic Energy Commission. One considered the evaluation of past production and the other an evaluation of the effectiveness of inspection and control procedures.² The latter document was prompted by discussions about whether or not International Atomic Energy Agency procedures could be effectively employed for disarmament purposes.

Our association with international safeguards through the Division of International Affairs began as a joint effort in 1959 with the Technical Branch of that Division to formulate an appropriate policy for determining the frequency of inspection. This study eventually served as a basis for the frequency formula included in INFCIRC-26,³ which was issued by the

* This paper is based on work performed under USAEC Contract AT(45-1)-1830.

IAEA on March 30, 1961. Following this, a series of training manuals on nuclear materials control, which has received wide distribution, was prepared in cooperation with Phillips Petroleum Company, Atomic Energy Division.⁴⁻⁷ More recently, three reports have been published in connection with the extension of IAEA safeguards. One evaluates safeguards procedures at the Yankee reactor (July 1965);⁸ the second considers safeguards at chemical-processing plants (September 1966);⁹ and the third discusses safeguards at fuel-fabrication plants (June 1967).¹⁰

Recently, at the request of the Division of Nuclear Materials Management and the Division of Regulation, problems in connection with the effectiveness of domestic safeguards have been considered.

Characteristics of Inspection and Control Systems

During the course of these studies a characterization of an inspection and control system has evolved as shown in Fig. 1. The basic elements of any safeguards system may be divided into containment, surveillance, and material accounting, or more properly, material balance accounting. Containment can involve the use of locks, tags, and seals, or it may involve physical restraints in the sense of posting of guards and/or other monitoring devices. Surveillance involves not only direct observation and monitoring, but equally importantly, the verification of plant operation and the certification of plant data. Material balance accounting is a basic tool of a control system and may not only involve records, reports, and source data obtained from the plant, but also independent agency measurements to the extent feasible and/or necessary to achieve a sufficiently independent material balance. Containment and, to some extent, surveillance provide direct evidence of diversion, i.e., they operate essentially on a go no-go basis. By contrast, material accounting procedures together with the observation and certification of plant operation, provide indirect or circumstantial evidence about whether or not a diversion has taken place. One of the important considerations in evaluating the effectiveness of safeguards is determining when action can or should be based on circumstantial or indirect evidence.

<u>Elements</u>	Containment		Surveillance		Material Accounting	
	Locks and Seals	Observation		Certification of Plant Data	Agency Measurements	Records and Reports
		Monitors	Direct			
<u>Actions</u>						
<u>Type of Evidence</u>	Direct			Indirect		

Fig. 1. Characteristics of Inspection and Control Systems.

Fig. 2. Systems Effectiveness.

An inspection or control system achieves effectiveness through its capability to detect diversion, and, directly or indirectly, to deter or prevent diversion attempts. As shown in Fig. 2, containment, probably the most effective way of deterring diversion, represents the most clear-cut case for evaluation; for if a diversion were attempted, broken locks or seals would clearly indicate the fact. However, there is an obvious difficulty here because effective containment limits the mobility of material or restricts access of plant personnel,

and this may be incompatible with the operation of the process. The fact that the neatly packaged fuel elements in reactors are almost immobile is the main reason why containment procedures provide effective reactor safeguards.

The second type of inspection activity, surveillance, can both detect and deter diversion. The effectiveness of surveillance, however, is very difficult to quantify. Surveillance will be more effective if conducted continuously during plant operation. Anyone planning a diversion that might be detected by surveillance will obviously make his attempt when surveillance is minimal or absent. Hence, any nonrandom or announced periodic surveillance effort would be completely ineffective against a planned diversion attempt.

Although it is very difficult to determine how a given level of surveillance affects the probability of a diversion attempt and/or the probability of detecting a given amount of diversion, it is possible to establish the relative effectiveness of different levels of surveillance. For example, if one inspector has a certain probability of observing a diversion, it is possible to compute the probability with which one of three inspectors acting independently would observe a diversion. By means of a subjective evaluation of a base level, computations of this type have been employed to obtain the relative levels of effectiveness in given types of facilities.

Containment and surveillance procedures detect a diversion now or one recently attempted. By contrast, the use of material accounting procedures determines indirectly whether or not diversion has taken place over some past period of time. Thus, it is possible through effective material accounting procedures to "catch up" with a diversion that has taken place at some time in the past. In this sense, the material balance portion of the inspection system can provide an upper limit to the amount which can

be diverted. On the other hand, the evidence provided by this type of inspection activity is always circumstantial unless after-the-fact investigation can provide direct evidence of diversion. There is also a lower limit to what can be detected, in the sense that some small (or perhaps not so small) amount of material can always be diverted without being detected by material accounting. Both the deterrent effect and the detection capability of material accounting are highly dependent on the amount and type of material involved.

The operation of the system is summarized schematically in Fig. 3.

Fig. 3. Control Actions and Results.

Effectiveness

When we consider the effectiveness of a given system as a function of complexity, we must consider both the characteristics of the plant itself and the environment in which it is located. The amount of effort required to achieve a given effectiveness or the effectiveness achieved with a given effort will vary with the size of the facility and the throughput and complexity of the process. Because the contained nuclear material is immobile, reactors in general present a safeguards problem that is different from that of either chemical-processing plants or fuel-fabrication plants. Furthermore, the highly radioactive materials encountered in chemical

processing make the problems of safeguarding chemical-processing plants different from those of fuel-fabrication plants.

Any evaluation of effectiveness must take into account the environment. Effective material accounting in a U.S. contractor facility, where adequate technical facilities are immediately available and where the AEC maintains complete records of all associated process inputs and outputs, is considerably easier than the inspection of an isolated quantity of safeguarded material in a foreign plant with the closest laboratory 3,000 miles away. Obviously the effort required to monitor the process and to establish a verifiable material balance will be markedly different in the two instances.

The effectiveness of any inspection will obviously vary as a function of the effort involved. In the first place, it will be highly dependent on the technical capability both of, and available to, the inspectors. It is also obvious that the effectiveness will vary with the number of personnel, a key point being whether or not the plant is under continuous observation. There has been much emphasis, and rightly so, on developing adequate instrumental means of measurement and appropriate analytical procedures which will provide an independent capability on the part of the inspectors.

Fig. 4. Dependence of Effectiveness on Complexity and Effort.

As a consequence of these considerations, the effectiveness of a given safeguards system is a function of the size, or complexity, of the facility and the magnitude of the inspection effort. Figure 4 shows an example of the relationship between these factors.² The general relationships in Fig. 4 are valid in most safeguards applications, although they may be difficult to quantify. It is a truism that no system will be 100 percent effective. It is equally true that there is no technical means for deciding just what combination of effort and effectiveness is right for a given facility. One of the difficulties in evaluating the necessity for resident inspection and the motivation for research and development has been the lack of a precise definition

of exactly how effective safeguards are required to be. At best we have been able to determine approximate relationships of the type shown in Fig. 4 so that a balance can be achieved between the effectiveness and the cost for a given facility.

As a final point we might consider the possibility that unless appreciable further work is done to improve present measurement methods and capabilities, safeguards capable of ensuring the necessary effectiveness

may not be feasible. Figure 5 shows the situation graphically. If a minimal inspection effort can achieve a material-balance precision that is better than that required for effective safeguards, then obviously such a minimal inspection is sufficient. However, if the complexity of the facility increases to the point where a minimal inspection effort cannot achieve a satisfactory precision, we must increase the inspection effort. We proceed along this line of constant precision (see Fig. 5) until we reach the point where the complexity and/or throughput of the facility are so great that even with a maximum effort in support of a material balance, it is not possible to achieve the necessary precision. If the required level of safeguards precision is to be maintained at these higher throughputs, it is necessary that surveillance and containment efforts supplement material-balance accounting. The relationship between size, complexity, and effort shown in Fig. 4 is really applicable only to the intermediate situation ($\sigma_s = \sigma_i$).

Fig. 5. Feasibility of Effective Safeguards.

A simplified explanation of Fig. 5 follows:

Let the Y-axis represent kilograms diverted, the X-axis kilograms of throughput, and the horizontal line labeled σ_s represent an "allowable" loss under safeguards control. Two levels of inspection intensity are shown. It is assumed that the potential diversion increases as the throughput increases (though not necessarily as a linear function).

Starting at low throughputs, safeguards requirements are met by audit inspection until the corresponding line crosses σ_s . Further increases in throughput require an increased inspection effort along the horizontal line $\sigma_s = \sigma_i$. Finally, at higher throughput, full inspection ($\sigma_a = \sigma_i$) by material accounting alone no longer is adequate. If σ_s is to be achieved at these throughputs, it is necessary that surveillance and containment efforts supplement material balance accounting.

Thus far the basic elements of a safeguards system and some of the problems encountered in evaluating its effectiveness have been described. From the viewpoint of research and development on safeguards, it is probably fair to say that the more important areas for further improvement lie in

- 1) containment and monitoring procedures that do not interfere with the primary purpose of the process,
- 2) improved analytical procedures, particularly those that are nondestructive,
- 3) the consideration of an interlocking system of reports that will enable effective cross-check control on a system rather than a given facility, and
- 4) a continuing study of the systematic evaluation of effectiveness.

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DR. McGONNAGLE: We will proceed to the next paper. Mr. R. P. Wischow is assistant general manager of Nuclear Fuel Services.

SAFEGUARDS PROCEDURES FOR A NUCLEAR FUEL REPROCESSING PLANT*

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(Paper presented by Mr. Wischow)

Nuclear Fuel Services, Inc. recently had the opportunity to participate in the development of a system that may be used to apply safeguards in general to a nuclear reactor fuel-reprocessing plant and specifically to the existing NFS West Valley facility. In this program it was desired to develop a set of procedures by which an existing chemical-reprocessing plant designed for the commercial recovery of plutonium and uranium could be adequately safeguarded by a cadre of inspectors while the plant was operating.

Part of the difficulty in developing any system of safeguards is that a generally accepted definition of the term "Safeguards" does not exist. As it has been used throughout this study, the term has had the following meaning:

Safeguards is the organization and system by which an assurance can be given that, if fissionable material is diverted from the normal and proper channels associated with the safeguarded facility, there is a high degree of probability that the diversion will be detected.

There are at least two direct approaches to provide such a system of safeguards, namely:

1. reliance upon the methods of accountability to determine independently that, within the limits of precision of the accountability program, the fissionable material has been accounted for satisfactorily, or
2. reliance upon methods of surveillance to permit observation of activities which may be suspect.

Neither of these approaches is aimed at preventing diversion except in a secondary sense that the activities may discourage a would-be diverter by convincing him his actions are more likely to be detected. Rather, these approaches are aimed at detecting that diversion has occurred.

*Work performed under USAEC Contract AT(38-1)-452.

Application of safeguards to a spent-fuel-reprocessing plant is not as clear-cut nor is it as straightforward as is the safeguarding of other nuclear facilities, such as a nuclear power plant. The difference is quite obvious in that from the time that the fuel enters the reactor complex until it is sent to the spent-fuel-reprocessing plant, there is essentially no method by which it can be diverted other than having the spent fuel assemblies sent to a clandestine or nonsafeguarded reprocessing plant. In addition, the physical facility of a power plant does not lend itself easily to a diversion of fuel elements from the assemblies. In contrast to this, a reprocessing plant accepts entire assemblies, subdivides these into small physical pieces, dissolves the material, and subsequently distributes the dissolved material into either an organic or aqueous media through miles of pipelines, a multitude of vessels with a multiplicity of recycle arrangements.

Over the last twenty years a number of studies were conducted, some of which are still classified, in which attempts were made to determine how to attack the problem of safeguarding a chemical-reprocessing plant. Considering the difficulty of the problem and the fact that these studies covered an extended period of time, during which the nuclear industry has undergone many rather drastic and remarkable changes, there is nevertheless a surprising amount of unanimity in these studies. In all cases, primary reliance was placed upon an audit of a system of the nuclear-materials accountability. In general, these systems also added some degree of physical or operational surveillance to the accountability determinations; however, surveillance was in general a subordinate effort with the primary effort being directed toward the accountability determinations. It is of interest to note that a unanimous conclusion of these studies was that no system of safeguards can be 100% effective.

Effectiveness is an extremely relative term which is applied somewhat indiscriminately to safeguards. Since it is not a simple matter to assign a quantitative value to the effectiveness of proposed but untested safeguards programs, the best estimate of the assurance is that the safeguards thus obtained approaches the limit of certainty of the accountability methods themselves. It is evident that, until a system of developed procedures is tested, only an estimate of the effectiveness as well as the amount of material verified by the inspectorate or defined as a percent of plutonium throughput can be alluded to.

It has been demonstrated that accountability programs are capable of accounting for $100 \pm 2\%$ of the plant throughput; therefore, one can conclude that it is possible to devise accountability monitoring or safeguards methods which independently assure that the accountability program of a safeguarded facility is as good as the facility claims it is. To this extent, such a program should have the capability to determine, with an assumed 95% confidence level, that a nondetected diversion of nuclear materials

would be less than the uncertainty of the accountability program. This, of course, leaves something to be desired, since even 2% of the throughput may be still a sizeable amount of fissionable material. For instance, processing fuel containing about 5 kg plutonium per tonne at a rate of 225 tonnes uranium per year would result in an uncertainty in the accountability of about 20 kg plutonium per year at a statistical uncertainty limit of 2%. Obviously, the entire 20 kg could not be diverted without a bias becoming apparent.

To discuss safeguards in its proper perspective, it is essential to establish basic ground rules or assumptions. The safeguards system as developed at NFS had several critical assumptions, namely,

1. The safeguards system shall be capable of detecting diversion of nuclear materials but shall not necessarily be capable of preventing diversion.
2. Safeguards will have the capability of establishing an independent nuclear material balance across the plant.
3. From receipt of spent-fuel shipments to the dissolvers, the plant accountability system is supplemented by physical security. From the feed accountability vessel to product loadout, primary reliance is on the inspector's independent material balance.

Other subordinate assumptions are necessary to define the details of the program and stipulate the requirements of the plant and inspectorate personnel, but these assumptions are not critical to the basic safeguards program.

It is of fundamental importance to clarify a major assumption, that is, for the present discussion, the safeguards development was directed toward an inspection of an isolated fuel-reprocessing plant and not a plant that was a part of an overall safeguarded fuel-cycle complex. The obvious conclusion is of course that many more people are required to safeguard an independent facility than would be required for the same facility that was a part of a fully safeguarded nuclear-fuel cycle. In the latter case, a true system of audit or check and balance could be effectively utilized to verify whether or not a diversion has occurred.

An effective safeguards program must be based on procedures that are primarily used to describe the plant accountability program and that would:

1. be relatively easy to implement and carry out;
2. be comprised of individual methods any one of which could be used or deleted at the discretion of the inspection manager;

3. not interfere unduly with the plant operations or functions;
4. be practical and as economical as possible, and still provide a requisite measure of effectiveness;
5. be based on the state-of-the-art and use essentially existing inspection concepts, that is, surveillance, examination, and verification.

Safeguards systems should provide an independent material balance that is at least as good as the plant system was capable of producing, that is, using the accountability system of the plant, the inspectorate could then independently verify the disposition of safeguarded fuel. In addition, surveillance can be used to increase the effectiveness beyond the normal limits of the accountability program. For example, the accountability program is nominally capable of accounting for $100 \pm 2\%$ of the safeguarded material and guaranteeing 98.5% recovery of the input material. Both of these values are based on a 95% confidence level. Therefore, it appears that a realistic and effective program with limited expenditures can be done by restricting the safeguards limits to those acknowledged by the plant accountability program. No attempt should be made to extend the planned limits to those beyond what the plant (in an honest, nondiversionary approach) is capable of doing. It is felt that such a program would not be unduly restrictive to a safeguards inspectorate with regard to people or capabilities as well as to the plant interference with process throughput. It is again emphasized that the limits suggested, that is, accountability alone, will to some extent be extended by virtue of the surveillance effort applied.

To provide a high degree of assurance that the location and disposition of safeguarded plutonium are known, an independent determination of the accountability, made by independent sampling and analysis by the inspector in addition to surveillance of the overall facility, should yield an effectiveness approaching or surpassing the accountability program as a limit of assurance against diversion. It is believed that to provide, implement, and utilize this extended program, it is essential to use television systems and other relatively sophisticated apparatus that may include monitors, alarm devices, tamper-proof seals, laboratory facilities, and analytical equipment. It is difficult to estimate, prior to a testing program, the detailed manpower requirements for such systems; however, as a preliminary estimate, a staff of 18 resident inspectors with the following capabilities appears realistic:

- Inspection Manager (1)
- Assistant Inspection Manager (2)
- Accountability Inspector (1)
- Analytical Inspector (4)
- Shift Inspector (10)

TABLE I. Estimated Annual Labor Costs of the Proposed Safeguards Program

Probable Limit of Safeguarding (% of plutonium throughput)	98
Manpower Cost (\$)	
Inspectorate	450,000
Plant Operator	33,600
	<hr/> 483,600

It is expected that such a program would cost roughly \$500,000 annually for an anticipated effectiveness of 98% (see Table I). Labor costs were based on the assumption that the program would be applied on a continuing basis, i.e., the entire plant is safeguarded continually for all fuel processed.

It is of interest to convert these values into unit costs based on the amount of material processed. As shown in Table II, the program translates into a cost approximately a little less than one-tenth the cost of reprocessing or about one-twentieth the value of the recovered plutonium and by comparison, is approximately the cost of waste storage including perpetual care.

TABLE II. Estimated Unit Costs for Safeguards Accountability Program

Labor Cost (\$/yr)	483,600
Plant Throughput (tonnes uranium/yr)	225
Labor Cost per tonne Uranium Processed (\$/tonne)	2,150
Plutonium Throughput (kg/yr) ^a	1,125
Equivalent Electrical Power (kWh/yr) ^b	3.2×10^{10}
Safeguards Cost (\$/g Pu)	0.43
Safeguards Cost (mill/kWh)	0.015

^aAt 5 kg Pu/tonne of fuel.

^bAt 20,000 MWd/tonne burnup and 30% conversion efficiency

However, it should be noted that even this program that approaches the limits of the plant accountability program would still leave an uncertainty of about 2% of material which lies outside the expected limits of the plant accountability program. At 2% of a throughput of 225 tonnes/yr, this uncertainty is about 20 kg of plutonium per year. To safeguard this specific 20 kg of plutonium would probably require a one-to-one ratio of inspectors to operators, since this concept would be attempting "100% effectiveness." Such a program may cost in excess of \$2,500,000 per year. To relate this cost to the additional 20 kg of plutonium which is safeguarded, the incremental \$2,000,000 would amount to \$100,000 per kg of plutonium. Even if the total cost were to be charged against the total throughput, the unit cost would be about \$2.50 per gram of plutonium or nearly one-fourth its current value of \$10.00 per gram. To this extent, it's questionable whether "total" safeguards is realistic.

If it is desired to extend the effectiveness of a safeguards program beyond the natural limits imposed by the statistical limitations of the accountability program, additional physical surveillance must be superimposed upon the operations in the plant. One method to accomplish this is to provide a sufficient number of people that they can be everywhere most of the

time and observe nearly everything that goes on. Such a program has numerous obstacles in addition to the fundamental cost aspect. Large numbers of observers will result in a significant interference with plant operations. As the number of inspectors increases, the more certain it is that severe personality conflicts will develop between the inspectorate and the plant management. This conclusion has been borne out by the few actual safeguards test situations which have been attempted. Finally, finding even a small cadre of inspectors with sufficient background to do their jobs adequately is going to be difficult enough without having to recruit a large number of qualified inspectors.

In summary, the whole matter of safeguards revolves around several basic points. A clear definition of safeguards is needed. In the program developed at NFS, it was assumed that it was desirable to determine whether diversion of fissionable material has occurred up to the limits of the accountability programs which can be applied to an undertaking of this type. Such a program can be implemented for a cost which seems modest in relation to the benefits to be derived. However, to extend the program to include all the fissionable material in a plant, even that which lies outside the limits of accountability, will be extremely costly in relation to the value of the material safeguarded. At the same time, it is necessary to establish some goal for the degree of effectiveness for safeguards. Closely tied to the degree of safeguards is its cost. It is of primary concern to determine at what point the increasing cost of additional assurance passes the point of diminishing return. In the course of testing the developed safeguards procedures, one can provide more and better data upon which to base this judgment, but in the final analysis an answer must be given as to how much this assurance is worth.

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ANALYTICAL MEASUREMENT METHODS AS APPLIED TO SAFEGUARDS

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Essentially the question in Safeguards is "How much uranium-235 or plutonium-239 and -241 is there in a certain facility?" In order to answer this question we first have to know how many gallons, liters, pounds, or kilograms of the material that contains the fissionable elements are in the inventory. The next problem is to obtain a representative sample of the material, and finally to determine by analysis the amount of the uranium, plutonium, or both present in grams per liter, grams per gram, or in some other concentration units.

In a discussion of chemical analysis, the terms precision and accuracy have to be defined. Precision is a measure of reproducibility of a method, whereas accuracy is a measure of correctness of a method. Accuracy is obtained by using the method for the analysis of a sample of similar composition in which the concentration of the element in question is known. The analyst is vitally interested in both, for he cannot achieve accuracy without precision. The recipient is interested in accuracy alone and only in accuracy sufficient for his purpose.

Let us now look at what can be expected insofar as accuracy and precision are concerned in the analysis of fissionable materials. In most chemical analyses the best precision and accuracy obtained is about 1 part in 1000 or 0.1%. This means that if we had a material actually containing 1000 g of uranium, the analysis would indicate it contained between 1001 and 999 g. In certain select cases one may be able to obtain a precision of one part in 2000 or 0.05%, but this is rare. Therefore, we cannot expect much better than 1 part per thousand. In some cases, however, the precision and accuracy may be much worse. Needless to say the highest precision and accuracy should be obtained in those areas where the greatest concentration of fissionable material occurs.

It is therefore necessary to have accurate methods of analysis. Since the cost of the analysis is also important, in certain instances some kind of a compromise may be necessary. However, since we are vitally interested in the safeguards problem, an accurate method of analysis may not necessarily be the cheapest one. In fact the opposite is generally true. Certain methods of analysis will not give the necessary accuracy. It is clear then that there is need of improvement in both methods of sampling and in methods of analysis.

In order to obtain an idea of problem areas let us consider the fuel cycle of certain fuels insofar as determination of the fissionable materials are concerned.

The feed used in preparing essentially all uranium fuel elements is UF_6 . No problems arise in obtaining a representative sample. This UF_6 is usually supplied with an isotopic analysis for the uranium-235 material. Chemical methods of analysis are simple. The purity of this material is such that it can be weighed on the stoichiometric basis for the uranium content with correction for minor impurities that may be present.

The UF_6 is converted to UO_2 and in some instances to metal for use in the preparation of alloys. Both the UO_2 and the uranium metal are readily sampled and analyzed. Up to this stage there is no problem in obtaining an accuracy of 1 part per thousand for the chemical assay of the material. In fact, with experienced laboratories a standard deviation of 0.01 to 0.02 is not uncommon in analyzing UF_6 . Table I gives an indication of the accuracy and precision obtained. The precision and accuracy with UO_2 is quite comparable with that obtained with UF_6 .

TABLE I. Variation (%) in Analysis of Uranium in UF_6

Laboratory:	A	B	C	D	E	Average	Theoretical
6 months	67.58	67.58	67.57	67.58	67.60	67.585	67.62
Standard Deviation	0.02	0.03	0.01	0.02	0.01		
6 months	67.60	67.59	67.58	67.57	67.60	67.59	67.62
Standard Deviation	0.01	0.004	0.01	0.004	0.02		

Since the uranium-235 content is of prime importance, it is necessary to obtain the concentration of this fissionable isotope. The precision obtained with the various methods are given in Table II. The accuracy is obtained by reference to a standard. At present, accepted standards are those uranium isotopic standards issued by the National Bureau of Standards. Certain gaseous-diffusion plants have also reliable standards.

As indicated in Table II, for safeguard purposes these methods appear to be adequate for the determination of the isotopic composition of uranium-235. The spectrochemical method may have problems when the content of uranium-236 is quite appreciable. Since essentially all these methods are applied to purified materials, the variations indicated apply to all types of uranium-bearing materials after separation of the uranium.

TABLE II. Methods for the Isotopic Determination of Uranium-235

Method	Coefficient of Variation		
	1-5%	20%	90+%
Mass Spectrometric			
Electron Bombardment (Gas)	0.04		0.01-0.03
Surface Ionization	0.5-1.5	0.1	0.01-0.03
Spectrochemical	0.2-10	2	0.4
Gamma Spectrometry	2-5	3	0.5
Neutron Activation	3	2	0.7

The cost of the isotopic analysis is in inverse proportion to the precision obtained, the more precise measurements requiring a considerable outlay in capital equipment. The method employing electron bombardment of gaseous UF_6 , however, requires a number of instruments to cover various ranges. Since the measurements are made against standards, the accuracy will depend on these standards. The bias from the true value should not be greater than the precision and should be satisfactory for safeguard measurements. Investigations of cheaper instrumental methods for determining isotopic composition could possibly be quite fruitful and may lead to methods especially useful for survey purposes.

The next group of materials to consider are fuels themselves. These may be coated metals, alloys, cermets, ceramics, coated carbides, etc. During fabrication it is possible to obtain a sample for analysis, but in many instances inhomogeneity is a troublesome problem.

The final fuel rods cannot be sampled, and it is in this field that non-destructive methods of determining the uranium-235 content are used. Although certain pieces of equipment, such as used at Los Alamos, enables one to obtain a precision and accuracy as good or better than chemical analyses on the same material, in many instances this is not the case. The entire fuel element is measured at one time at Los Alamos; however, it is necessary to have a fuel element of similar composition to use as a standard.

Results on a $\text{UO}_2\text{-CaCO}_3\text{-ZrO}_2$ ceramic fuel are given in Table III. This table shows a comparison of results between two laboratories and indicates what might be expected from the analytical standpoint.

At this time it is well to consider scrap from fuel-element fabrication. Essentially in the same category but with greatly increased problems are the samples resulting from the processing of spent fuel.

TABLE III. UO_2 Content (%) of Ceramic Fuel Element

Sample	Calculated UO_2	UO_2 Found (Lab 1)	UO_2 Found (Lab 2)
1	54.9	54.91	55.11
2	54.9	55.43	55.50
3	54.9	54.76	54.91
4	40.2	40.25	40.50
5	40.2	40.31	40.48
6	40.2	40.28	40.34
7	26.5	26.90	26.95
8	26.5	26.90	27.08
9	26.5	26.54	26.76

In order to obtain a representative sample of cold uranium scrap, it is necessary to dissolve a certain lot or lots of the scrap and take a sample of the solution of the dissolved material. When complete solution is not obtained, the residue remaining leads to problems in determining the uranium content. Certain counting techniques used to determine the uranium-235 content of these residues have not proven entirely satisfactory. When the various components of fuels are considered, many of which are notorious for causing problems in analytical work, it does not come as a surprise that the agreement in the analyses of solutions from fuels is not as good as with the feeds. It is in this area of scrap reprocessing and spent-fuel processing that the biggest problems in safeguards may occur.

To obtain an indication of the precision and accuracy in the analysis of uranium-scrap solution, a series of solutions of a composition similar to those found in a recovery plant were made with known amounts of uranium. Different types of feed were simulated. The results are shown in Table IV. It is quite evident that the precision and accuracy with scrap solutions is entirely different from that obtained with feed materials.

TABLE IV. Scrap-recovery Analysis

Laboratory	Average Difference (%) from Prepared Value (Relative)					
	UO_2 -SS	U-Mo	U-Al	BeO- UO_2	U-Zr-Mo	U-Nb
A	+0.009	-0.007	+0.40	-0.06	+0.53	-0.03
B	-0.21	-0.35	-0.0003	-0.35	-0.42	+0.33
C	-1.79	+0.29	-1.19	-1.18	-1.47	-0.22
D	-0.12	-0.66	+1.64	-0.53	-1.54	-0.18
E	+0.23	-0.83	+0.40	-0.30	+0.13	+0.04
F	+0.96	+0.14	+2.77	-0.16	-2.06	-0.25
G		-1.4	+0.88		-0.44	
H	-0.12		+0.12			-0.22
Range of Prepared Value, g/kg	4-6	9-10	2-8	4-7	5	18

A comparison of the values obtained on scrap solutions that have been refereed is given in Table V. The data are quite indicative of the variations in results found with scrap material.

TABLE V Enriched Uranium Scrap Solutions Referee Samples,
Uranium in Grams/Kilogram

Sample No	Lab A	Lab B	Referee	Difference, %
1	0 93	0 91	0 98	-2 15
2	0 94	0 89	0 96	-5 32
3	23 07	22 43	22 43	-1 56
4	23 31	22 82	23 27	-2 77
5	29 09	28 56	29 17	-1 82
6	24 59	24 11	24 58	-1 95
7	24 11	23 60	24 24	-2 13
8	24 00	23 44	23 92	-2 33
9	11 15	11 04	11 14	-0 99
10	12 88	12 75	12 90	-1 01
11	13 62	13 44	13 56	-1 32
12	20 81	20 43	20 76	-1 83
13	5 53	5 38	5 48	-2 71

Although these methods and results may be adequate for "inhouse" requirements, they may not be satisfactory where safeguards are considered. Note that the results are expressed as g/g; it has been found that results are more satisfactory on a weight basis of these solutions rather than on a volume basis.

The press-cake analyses from scrap have varied considerably, as indicated in Table VI. As can be seen there is a definite trend to positive bias on the basis of Laboratory A. If any considerable amount of press cake

TABLE VI Uranium in Press Cake

Lab A, g/g	Lab B, g/g	Difference, %	Relative to A, %
11 87	8 14	+3 73	+31 4
3 97	2 63	+1 34	+34
5 02	2 94	+2.08	+41
2 90	1 87	+1 03	+36
1 60	4 87	-3 27	-20
1 50	0 12	+1.38	+92
25 36	1 74	+23 62	+93
22.49	22 54	-0 05	-0 2
2.27	2.36	-0 09	-4.0
2.96	2 46	+0 50	+17
16.80	13 24	+3.56	+21
17.36	15 39	+1 97	+11
16.77	15.92	+0 85	+5
0 69	0 75	-0 06	-9
18 15	17 1	+1 05	+5 8

is involved, a series of discrepancies could occur. In some instances as much as 2 kg have been found in the residue from a solution process.

The processing of irradiated reactor materials has presented special problems both in sampling and analysis. Interchange of samples is extremely difficult and interchange results have been rarely available. Methods employed have to be applicable to the determination of small amounts of the element in question, since usually small samples are taken because of the radioactivity of the solutions. Since the solutions are very radioactive, the fissionable materials must be either separated or a method used that is not affected by the radioactivity.

An isotope dilution technique employs a small sample and is used to a great extent for spent fuel analysis. The precision is about 1 to 2%.

Exchange programs with dissolver solutions from spent reactor fuel will, I believe, be a program to consider in the future.

There are many places, such as in effluents, where it is necessary to use methods that are applicable to small amounts of uranium. In such cases colorimetric or fluorometric methods are usually employed. The colorimetric methods are more precise and accurate than are the fluorometric. In some instances the amount of uranium is extremely small and the cheaper fluorometric method may be entirely satisfactory. The fluorometric method is more of an art, however, than a science, and the precision and accuracy may depend to a large extent on the operator. Results have been reported for which the precision may range from 1 to 20% of the amount present. Possible improvement in instrumental methods could make this an "in-line" operation.

In regard to plutonium analyses, as with uranium let us consider the feed to be used in the preparation of plutonium fuels. The feeds are generally plutonium nitrate, plutonium metal, or plutonium oxide. At the present time plutonium nitrate is also returned to the Atomic Energy Commission from processing of irradiated fuel. Plutonium nitrate solutions that have been aged can be readily shipped. Table VII shows variations

TABLE VII Plutonium in Known Plutonium Nitrate Solutions
Deviation (%) from Prepared Values (126-133 g/kg Soln)

Month	A	B	C	D	E	F	G	H	J	K	L
August	+0.05	+0.41	-0.29	na	-0.22	-0.86	-0.36				
September	-0.01	+0.78	-0.19	+0.16	-0.38	+0.62	nr				
November	+0.08	-0.13	-0.24	+0.18	+0.13	+0.57	-0.13				
February	+0.01	+10.1	-0.27	+0.72	+0.01	+1.83	+0.33	+0.47	+0.01	-0.15	+0.17
March	0.00	+0.40	-0.29	-0.06	+0.51	+0.56	-0.22	-0.53	+0.40	-0.06	+0.95
May	+0.07	+8.76	-0.26	-0.34	-1.18	-0.04	-0.87		-1.32		+0.04
Plutonium in Process Samples (6 months). Coefficient of Variation (%) from Individual Laboratories											
	0.34	0.34	0.54	0.18	0.73	0.47	0.24	0.18 ^a	0.39 ^a	0.29 ^a	0.32 ^a

^a2 to 3 months

from prepared values of plutonium nitrate solutions analyzed by essentially every laboratory of any importance in plutonium analysis in the United States. The precisions of the plutonium determinations on process solutions are also shown in the same table.

As can be seen the precision and accuracy with plutonium nitrate solution is not as good as with UF_6 . Plutonium metal is usually assayed by difference after measuring impurities. When it is necessary to assay directly, the precision and accuracy is expected to be 0.2 to 0.5%. Plutonium oxide is a material that may change in weight, depending on the temperature of ignition. This is an important factor, and the variations obtained, therefore, may not be due to method variation but to other factors.

The isotopic analysis of plutonium for plutonium-239 and plutonium-241, the fissionable isotopes, is invariably done by a mass-spectrometric method. The precision and accuracy based on a National Bureau of Standards standard for the isotopic analysis of plutonium are the same order of magnitude as for the analysis of uranium. In addition, the possibility of degrading the fissionable plutonium is not as readily done as with uranium. Certain instances have been known where uranium-235 of 93+% has been degraded. For safeguards it is important to check on this degrading, since only isotopic results can detect material in which natural uranium was substituted for enriched uranium. It is charitable to believe that all changes in uranium-235 have been due to human errors, but the possibility of diversion exists.

The fuels prepared from the above feeds present the same problem as with uranium except in the case of the nondestructive testing of plutonium results may not be as good as with uranium. Variation in methods of preparation cause variations in the homogeneity of fuels. This is true of alloys, ceramics and cermets. With high fired PuO_2 the stability and homogeneity are much better than with the above fuels. Again the methods of analysis are not the primary cause of variation in results. Most of the chemical methods are accurate to within 0.3 to 1%. There is also a tendency for certain laboratories to show a bias either positive or negative. Changes in the methods of analysis to improve the precision and accuracy as well as to decrease the costs of analysis should be quite fruitful. Plans to have exchange programs with various plutonium fuels are in progress. It is only from such exchange programs that answers as to reliability of analytical results can be obtained.

The processing of plutonium scrap is relatively recent. At present the processing of slag and crucible scrap, incinerator ash, power reactor fuel scrap and hood waste have had many shipper-receiver differences, in some instances being in the order of 20%. For single measurements of scrap whether by the use of instruments or by chemical analysis the reliability has not been much better than 25%. One can see that safeguards problems with plutonium scrap are major ones.

One can readily see that the scrap-recovery operations both for uranium and plutonium are troublesome areas as far as safeguards are concerned. Additional exchange programs with the newer plutonium fuels as well as attempts to solve the scrap analysis problem appear to be the major courses of action. The preparation of analyzed materials for use by all concerned in problem areas is to be recommended.

Little has been given on problems associated with processing of hot reactor fuels. Exchange information is not available except in limited cases, and this phase of safeguards warrants a special investigation of its own. Apparently analytical methods employed will give 0.5 to 2% for accuracy and precision.

Investigations leading to the development of instrumental methods of analysis, based on either radiochemical, chemical, or electrochemical operations in which results of analysis can be obtained from portable instruments, could prove fruitful.

With this brief survey I would like to close giving references to a few compilations of methods that may be of value. In addition to the laboratory manuals of the various contractor laboratories, the following may be considered:

1. R. Jones, Editor, Selected Measurement Methods for Plutonium and Uranium in the Nuclear Fuel Cycle, USAEC Report TID-7029 (Oct 1963).
2. C. J. Rodden, Contributing Editor, Analysis of Essential Nuclear Reactor Materials, USAEC, Superintendent of Documents, Washington, D.C.
3. Treatise on Analytical Chemistry, Part II, Edited by I. M. Kolthoff and P. J. Elving, Analytical Chemistry of the Elements, Vol. 9; "Uranium," Glenn L. Booman and James E. Rein, pp. 1-188, and "The Trans-uranium Actinide Elements," Charles F. Metz and Glenn R. Waterbury, pp. 189-440, Interscience Publishers, New York (1962).
4. Referee Methods for the Chemical Analysis of Nuclear Fuels, American Standards Association, Inc., New York, September 20, 1965, ASA N5.7-1965.

USE OF REACTOR CALCULATIONS AS APPLIED TO SAFEGUARDS

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INTRODUCTION

With the rapid growth and size increase of the new nuclear power-generating stations, the ability to predict fuel inventory in discharged fuel elements accurately is of prime concern. This is due to the fact that the discharge fuel from a power reactor is not processed immediately, and the value of this fuel must be established on the basis of computed fuel composition for an interim settlement. Since large amounts of fuel will be involved, high-degree accuracies in the computed isotopic content are necessary to prevent large discrepancies in the interim settlement. The accuracies of any calculational model depend, of course, on the basic methods and neutron microscopic-cross-section data used to perform the computations and on the interpretation of the reactor operational data which is used to determine the total production of energy in the core. The effect of each parameter on the accuracy of predicted concentrations is very difficult to identify.

In order to demonstrate the large amount of plutonium which will be produced by the second-generation types of reactors which are now in various stages of design and construction, Table I lists the approximate total core loadings and the approximate total plutonium-production rates for the U.S. reactors by startup year.

TABLE I. Approximate Core Loadings and Plutonium Production in
New U.S. Reactors by Startup Year

Startup Year	Approximate Total Core Loading of UO ₂ (kg)	Approximate Total Plutonium Production (kg/yr)
1967	250,000	407
1968	120,000	120
1969	551,000	810
1970	1,288,000	1790
1971	1,697,000	2820
1972	1,093,000	2070
1973	488,000	630

In reactor design, calculations of fuel depletion are performed in order to determine: (1) that the available excess reactivity will permit the core to operate in such a manner so as to achieve the design fuel exposures, and (2) that the hottest spot in the core will not exceed the design criteria at any time during core life. The computed isotopic concentrations obtained from these analyses are not of prime interest in so

far as fuel accounting is concerned, since the actual mode of reactor operation, in general, is different from that envisioned in the design period. Hence, fuel-accounting calculations should be performed during and after reactor operation in order that the actual reactor operating data, such as measured control-rod positions and total energy production, can be utilized in the computational routines.

This paper presents the results of work performed for the U.S. Atomic Energy Commission under Contracts AT(38-1)-207 and AT(38-1)-428 and their relationship in determining the feasibility of using reactor calculations as a means to material safeguards. The main tasks included under these contracts were: (1) an investigation of the feasibility of developing a simple method for use as a fuel-accounting tool, (2) an evaluation of the accuracies which are obtainable with this method, and (3) the setup of the method for seventeen specific reactors. The primary emphasis here is on the accuracies obtained with the method and the means to their improvement.

CALCULATIONAL TECHNIQUES

The calculational method selected for use as a fuel-accounting tool is denoted as the ISOCHECK Method. Its selection was based on the results of the initial two phases of the burnup study, called BUCAL,^{1,2} and it represents an effective compromise between accuracy of results and simplicity of calculation for the determination of heavy-isotope content in operating fuel elements of power reactors.

The application of the ISOCHECK Method to a specific reactor consists² of:

- 1) Calculation of assemblywise exposures with the FLARE Code³ for successive intervals of power operation using the known location and prior accumulated exposures of the fuel elements, the recorded average control-rod positions, and the observed thermal-power output history during the interval being analyzed. The FLARE Code computes the three dimensional power and exposure distributions using an effective one-group nodal model. The use of FLARE is essential for computational economy in analyzing boiling water reactors because of the interdependence of the power and void distributions and the ease with which FLARE handles this problem.
- 2) Determination of assemblywise isotopic contents by applying the FLARE-calculated exposures to prepared tables of isotopic composition versus fuel exposure.

The steps required for the setup and use of the ISOCHECK Method for a given power reactor are shown in Fig. 1. To begin with, all the necessary data for the reactor core design and operating conditions must be obtained from the reactors designers or operators. This includes the

Fig. 1. Flow Chart Defining Steps Required for Setup and Use of the ISOCHECK Method.

basic fuel-rod and fuel-element dimensions and material compositions, the control-rod description, the operating temperatures, and the power level. The information is reduced into appropriate forms for input into calculation codes, which are utilized to generate the few-group effective microscopic cross sections and the basic core constants. At present, the FORM Code is used for the above calculations of thermal-neutron spectrum, and the TEMPEST Code is used for the thermal group. CEPTRE is a P-3 mono-energetic transport code which is used to compute the thermal-group disadvantage factors. These are combined with the spectral-averaged thermal microscopic cross sections to obtain the effective thermal-group cross sections. The output from these codes is then inputted into a few-group, two-dimensional diffusion code, such as PDQ05, for cell calculations which yield the beginning-of-life values of the infinite multiplication factor k_{∞} and the migration area M^2 for each fuel type present in the core. These calculations are performed for the rodded and unrodded operating conditions and for three coolant void levels, if it is a boiling water reactor.

The effective microscopic-cross-sectional data are also used in conjunction with homogenized fuel-element number densities in a one-point depletion code to determine the average heavy-isotope composition of each fuel type as a function of fuel exposure. This is done for three void levels for boiling water reactors. The results are used to prepare the required tables of isotopic composition versus fuel exposure for each fuel type. Included in the results of the depletion code is the exposure dependence of k_{∞} .

In addition, the few-group diffusion code is also used to obtain the assemblywise power distribution at beginning-of-life operating conditions for typical control-rod patterns; in these calculations, the various regions of the core are represented discretely. These results form the basis for obtaining the values of the adjustable parameters used by the FLARE Code. This is done by inputting the previously calculated appropriate cell k_{∞} and M^2 values into FLARE and varying the reflector albedos, the transport kernel, and the transport kernel mixing parameter until the FLARE results match the diffusion calculation. These adjustable parameters are then fixed for use throughout the burnup cycle.

The basic input into FLARE requires k_{∞} and M^2 for each fuel type as a function of exposure, voids, and amount of control; the basic fuel-element dimensions; the core-loading and orificing patterns; and parameters used to fit the curve of void level versus quality.

Once the fundamental FLARE input has been generated and the isotopic tables have been prepared, the ISOCHECK Method is essentially ready to be used for the determination of the isotopic content in the various fuel elements. If the reactor has been operated prior to the setup of ISOCHECK it is necessary to obtain the previous operating history and select a starting time for the beginning of the calculations. The estimated assemblywise exposures prevailing at this starting time must be inputted into FLARE. The

ISOCHECK Method then follows further operation of the core to the end of a cycle. The FLARE results are then combined with the isotopic tables to determine the heavy-isotope content in each of the assemblies.

The use of FLARE in the ISOCHECK Method makes the method relatively easy to use, but it may lead to compromises in core representation, since FLARE is not sufficiently flexible to cope with the many different designs of developmental reactors. In general, though, the new type of reactors which are being designed and built do not have these geometrical complexities, and therefore the application of ISOCHECK to them is straightforward. The main problem in the use of the ISOCHECK Method lies in the fact that either new fuel types may be loaded into the core or core modifications may take place which influence the physics characteristics at the end of each cycle. In these instances, it is necessary to go back and recompute physics parameters in order to update the method; however, this is an inherent disadvantage in any calculational tool used for fuel accounting. But in general, the feed fuel for the new reactors will not change once an equilibrium cycle has been obtained.

PRESENT ACCURACIES

The present accuracies which are obtainable with the use of the ISOCHECK Method have been established from the results of the initial phase of the BUCAL work,¹ and they depend explicitly on the reactor type and on the type of fuel used in the core. These accuracies are considered to be characteristic of the state-of-the-art of reactor physics at this time. For light-water-moderated cores which are fueled with UO_2 enriched in U^{235} , the accuracies shown in Table II can be expected. As noted, the accuracies will increase as the batch size increases, because of the decrease in the uncertainty of the computed batch exposure. Also, there is a slight increase in accuracy as the batch exposure increases for the same reason.

TABLE II. Estimated Overall Uncertainties in Predicted Isotopic Compositions for Light-water-moderated, Enriched UO_2 Systems

	Total Uncertainty (%) in			
	U ²³⁵ Depletion		Total	
	MWD/MTU		Plutonium Content	
	10,000	20,000	10,000	20,000
Large Batch (≥ 100 Elements)	3.2	2.8	7.3	7.1
Small Batch (~ 20 Elements)	5.0	4.4	7.9	7.3

For cores containing ThO_2 enriched in U^{235} , the accuracies are $\pm 10\%$ for U^{233} , $\pm 7\%$ for U^{235} , and $\pm 7\%$ for total fissile uranium. These latter accuracies are applicable only for batches of fuel elements which contain

about 20 elements. All the accuracies quoted here are only for cores in which geometrical complexities are absent.

The accuracies are based on the combined uncertainties associated with the determination of the batch exposure and with the calculation of the conversion ratio and isotopic cross sections at constant exposure. These uncertainties were determined by comparisons of calculations and measurements for several different cores, but the primary data pertained to Yankee Core I. In this latter comparison, data of isotopic content were only available for individual fuel pellets and the batchwise uncertainties were obtained by a simple averaging procedure.

CONCLUSIONS

As evidenced by the previously quoted accuracies, utilization of a calculational method such as ISOCHECK for materials safeguards is not sufficiently conclusive, since $\pm 8\%$ of the total plutonium production and $\pm 5\%$ of the U^{235} depletion for nuclear fuel utilized in reactors of this country alone in the near future will represent very significant amounts of fissile fuel. Uncertainties exist in the quoted accuracy values due to the lack of measured isotopic content in batches of spent fuel elements for comparison with calculations. The ISOCHECK Method has been set up for seventeen specific reactors.⁴⁻²⁰ The U.S. Atomic Energy Commission is presently engaged in obtaining results with ISOCHECK for the Yankee and Dresden-I reactors. Fuel elements from these reactors have been sent to the Nuclear Fuel Services facility at West Valley, New York, for reprocessing. The measured isotopic concentrations of fuel batches of each core will be checked against the results obtained with the ISOCHECK Method. At that time it will become possible to establish firmly the accuracies of the ISOCHECK Method. If these eventual accuracies of the method are deemed to be insufficient for fuel accounting and material safeguards purposes, it will be necessary to modify the multigroup cross-section sets used for computing the basic core constants and the effective microscopic cross sections in an effort to arrive at accuracies which are satisfactory. Use of comparisons of measured and calculated values for batches from various cores will assist in determining a cross-section set which will consistently result in the best possible overall agreement with experiments. Hence, in the near future, it will be possible to use reactor calculations for the purpose of material safeguards.

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DISCUSSION

DR. KINDERMAN: Thank you. Are there questions?

DR. WILKINS: I wonder if you would care to comment on the extent to which these uncertainties could be reduced if the computing industry would get on the ball and let us do more sophisticated calculations. The FLARE Code, for example, is not a multigroup, three-dimensional calculation.

DR. KERN: The problem is not so much with the computed distributions, because all we are really interested in essentially is the batch-wise exposure values. Even FLARE will do fairly well for such a calculation. Probably the main problem in the computations is in the codes which are used to calculate the effective cross sections, the spectra and the like nature.

DR. KOUTS: Suppose you were to do a complete reactor by this means; what would your uncertainty be?

DR. KERN: Are you talking about from the beginning of life?

DR. KOUTS: No, to 10,000 MW. You have large batches. What would it be for a complete reaction?

DR. KERN: Here again it is probably about half of that. As with larger batches, accuracies go down. Asymptotically, for a number of assemblies you would probably have an accuracy of about half of the 5 per cent that you have for small batches.

DR. KOUTS: I think you are probably a little optimistic.

DR. KERN: It may be.

DR. KOUTS: I think the uncertainty in heat balance and the uncertainty of direct burning of U^{238} will probably account for that much loss.

DR. KERN: That is as much as I was accounting for, as a matter of fact.

DR. KINDERMAN: Do these uncertainties reflect any uncertainty in the total quantity for isotopic content of the fuel charged? One hopes that one charges 100 tons of uranium in a reactor, but one may charge 99.5, and one hopes that the enrichment is 3 per cent, but it may be 2.95.

DR. KERN: I think this is probably for the total that you would charge to the reactor. It takes into account no other uncertainties.

CLANDESTINE DIVERSION OF SOURCE NUCLEAR MATERIALS IN A POWER REACTOR

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I am delighted to have the opportunity to participate in this Symposium, not only because of the impressive list of distinguished participants, but also because this Symposium is a manifestation of a spreading of the already great interest and concern at the policymaking level in the development of more effective systems of safeguards.

This spreading interest and concern has served to stimulate research on safeguards problems--some results of which have been summarized by previous speakers--but the principal thrust of this research has been essentially in the direction of increasing our data-gathering and interpretation capabilities. In the course of reviewing our present and prospective capabilities to detect diversion from reactors, I intend to raise questions concerning the incentives for diversion, the returns from limiting diversion opportunities, and the appropriate level of costs that we should be willing to incur for various systems.

Predictive methods and techniques of nondestructive measurement in the past have been subject to sufficient inaccuracies that the detection of diversion by cross-checking reprocessing results would have been subject to considerable uncertainty. With the exception of highly enriched uranium, all of the materials of interest are formed through neutron capture, so that a wide variety of materials and methods is potentially available to a would-be diverter. The possible materials include plutonium, U^{233} , and tritium. The methods would include variants of diversion of a part of the fuel normally used in the reactor; or diversion of material inserted into the reactor in addition to the normal fuel loading; or diversion of neutrons that would ordinarily be lost through leakage or through nonproductive captures; or, of course, a combination of these methods. The basic reliance in any potential method is on the uncertainty in predicting reactor performance.

Against this variety of possible methods, the detection system consists of visual inspection of reactor internals and fuel elements, theoretical calculations of burnup, and nondestructive-testing measurements. The theoretical calculations of burnup and the scanning results can then be compared to reprocessing results when the latter become available. Unfortunately, the reactor calculations and the nondestructive-testing calculations are still somewhat lacking in precision, and the reprocessing results are available only for a batch of fuel elements. The reactor operating data, the reprocessing data, and the data for initial fuel-enrichment composition are all

subject to errors or to deliberate misstatement, as the inputs are supplied by parties that must prudently be deemed adversaries. Moreover, there is as yet no indication that reprocessors are willing to be judged by what the computer says should be in the fuel rather than what is actually found in the initial assay.

Several potential methods of diversion have already been identified and safeguards countermeasures to each such scheme have been developed, but the basic problem is that no one knows for certain whether some other diversion methods may have been overlooked. I assume that bank examiners regard their methods as relatively foolproof, and yet every now and again another embezzler is tripped up--often more by accident than by the functioning of the examination system. Of course, the embezzlement problem is somewhat less complex, both because the material diverted--money--is readily measurable, and because the embezzlement conspiracy is usually one person or a few individuals, and does not involve officials of the bank. Even so, we have little idea how many embezzlers are currently operating undetected.

The moral to be drawn from this allusion is that no system of reactor safeguards can be considered truly foolproof unless measurement and prediction inaccuracies are eliminated--and that is only a necessary, not a sufficient, condition.

However, I would like to suggest that a foolproof reactor safeguards system, although desirable, may not be necessary when the incentives for diversion are considered.

In his letter of invitation to this conference, Dr. Seaborg states that "... plutonium will be produced in nuclear power reactors throughout the world by 1980--if our projections are correct--at the rate of more than 100 kg a day." Although I am more than a little skeptical of the bases on which such forecasts have been developed, it is evident that a diversion of as little as 1 percent of even a smaller production rate of plutonium accumulates a substantial stock of material. Nevertheless, the mechanical application of percentages to global production figures overstates the nature of the problem. Dr. Seaborg more nearly delineated the problem when he indicated our aim would be to develop methods "with a precision and accuracy sufficient to ascertain whether strategic quantities of nuclear materials could have been diverted to military uses." The operative word I would judge to be "strategic."

On this basis, the problem of safeguards in the nuclear nations--the U.S., the U.K., and France--may be viewed as of secondary interest, largely to be handled by disincentive systems such as those proposed in the Lumb Report. This is merely a recognition that, so far as reactors are concerned,

more stringent provisions may be needed to guard against possible diversion by a well-organized group operating with tacit or implicit government or military support than are needed to guard against a lesser conspiracy of a few individuals motivated by personal gain and operating contrary to the desires of government and the military. This proposition is largely due to the inconvenient form of material diverted from power reactors, and would be much more dubious in regard to reprocessing plants.

Among the remaining Nth countries outside the Sino-Soviet bloc, the most highly industrialized are distributed around the periphery of the Soviet Union itself. To satisfy the requirements for a strategic force to deter so powerful an adversary calls for a highly ambitious weapons program, as the British and French have belatedly discovered. That ambitious program would have to include an extensive effort of materials acquisition. As an example, consider the case of West Germany, for which the Arthur D. Little "high" forecast assumes an installed capacity of 9,000 MW by 1975. At a production rate of 0.2 kg plutonium per megawatt-year, reasonably typical of values given the present burnup projections, the annual production of this program would be about 1800 kg of plutonium. Even if it were possible to divert as much as 5 percent per year, this would represent no more than 90 kg diverted to weapons production. At the traditional estimate of 5-10 kg per weapon, this is sufficient for fewer than a score of nominal weapons per year. This rate would be so out of proportion to any reasonable estimate of West German strategic requirements, that a program based on clandestine diversion from reactors would be measured in decades. This, of course, abstracts from the further problems of covert reprocessing, weapons design and delivery, and the like. Since the forecast of German capacity is the largest of all the Nth countries, the above calculation represents an upper limit to the near-term potential for diversion.

Although the ultimate capability of the Chinese cannot yet be foreseen, the above assessment might well apply to a potential Japanese or Indian weapons program designed to serve as a deterrent to China.

This leaves for consideration a small group of countries involved in various regional disputes. These third-area conflict situations, by the very nature of the existing armed forces, are such that one or a very few nuclear weapons on either side would constitute a considerable strategic force. We have just staggered through an exceedingly trying period in the Middle East, and it is clear that possession of nuclear weapons by any of the parties to that conflict would have made that crisis far more dangerous.

Even for a country in a third-area confrontation, however, clandestine diversion of strategic amounts of material still requires an installed nuclear capacity large in relation to present generating capacities. A capacity of 500 MW would produce annually some 100 kg of plutonium; again assuming that as much as 5 percent could be diverted without detection, this

amounts to only 5 kg per year, barely at the threshold of usefulness. The only third-area conflict country with plans at present for capacity of this magnitude is India, and my somewhat heretical view is that prospects for the spread of nuclear power plants in third-area countries are not good, given a thorough economic analysis. In fact, I have yet to review a proposed nuclear project for an underdeveloped country in which the nuclear alternative should have been preferred on competitive economic grounds. This, then, suggests that to the extent we are able to control the urge to assist a country's economic development by means of special subsidization of nuclear projects, third-area safeguards problems are unlikely to become markedly more complex or extensive than they are at present. Moreover, the technological backwardness of most of the third-area countries suggests that they could not develop a nuclear materials and weapons capability without substantial outside assistance.

This brings me to a final topic which, like an hereditary defect in the family line, is mentioned only in a whisper, if discussed at all. This topic is the open diversion of a complete fuel loading. We have noted that a capacity of 500 MW produces roughly 100 kg of plutonium per year, but the amount in the complete core would be still larger. Such an amount clearly would be of strategic importance to a third-area country; indeed, even smaller installed capacities would have sizeable inventories of plutonium. Moreover, in contrast to the degree of leverage that can be exerted against countries using enriched uranium, there would be little direct cost associated with total diversion from natural uranium systems. This, of course, makes the natural uranium system attractive as an option-preserving strategy, regardless of the small economic penalty thereby incurred.

My purpose in raising this unpleasant prospect is to point out that clandestine diversion is not the only alternative available to a country, and that a country which carefully evaluates its potential future need for nuclear materials might well regard the slow and anxious process of clandestine diversion and stockpiling as inferior to merely maintaining a ready source of materials.

I have repeatedly used a 5 percent diversion rate in my examples above to suggest that even with this large a leakage, the accumulation of materials in strategic quantities is painfully slow. I am inclined to believe that a relatively uncomplicated and inexpensive reactor safeguards system using little more than present material balance and inspection/verification techniques is at least this accurate, and probably better.

By way of summarizing the assertions and implications of viewing the reactor safeguards problem in this way, we may note the following points.

First, the current safeguards methods lack the necessary discrimination to determine, in the absence of direct evidence, that small diversions from power reactors have occurred. Furthermore, there is no assurance that all of the potential methods of diversion have been identified and countered.

Second, in contrast to the slow and risky course of covert diversion, an alternative solution to the problem of materials acquisition might well be the open renunciation of safeguards and diversion of a complete core loading at a future date. These two points taken together suggest that the incremental benefits from marginal improvements in the safeguards system will not be large, so that the cost we should be prepared to incur to gain that improvement would be small. There is not much point in paying for redundant locks on the front barn door if the door in the back of the barn is to be left wide open.

Third, the accumulation of strategic quantities of nuclear materials through clandestine means appears unattractive (and possibly infeasible) for any country which wishes a nuclear force to deter the Soviet Union or, probably, China. This means that the incentive for clandestine diversion from reactors is likely to be greatest for countries involved in third-area conflicts.

Fourth, the evaluation of the differential incentives and strategic requirements among countries suggests that the most cost-effective application of scarce safeguards resources--and anyone who has reviewed the IAEA's budget will recognize the aptness of that description--is through a discriminatory system rather than one which concentrates equal effort on similar reactor facilities in all countries. This would free resources for safeguards functions at other facilities than power reactors and for research on some of the other pressing problems in efforts to limit the spread of nuclear weapons.

Finally, this analysis depends on each Nth country perceiving in an accurate and unambiguous manner that its strategic requirements would be poorly served by the mere possession of a few critical masses, however obtained. This will require a change in attitude in certain areas, wherein much of the dialogue has tended toward handwringing and other unhelpful reactions that can only reinforce an Nth country's belief that the possession of nuclear weapons in however small numbers is a desirable goal. We need much more forcefully and openly to set forth factual information about the enormous costs--monetary and otherwise--associated with attempts to secure nuclear weapons. We must endeavor to make plain to Nth countries that nuclear weapons, after all, are not some sort of treat that the big powers are trying to keep out of their grasp, but rather are a powerful drug that may produce dangerous and unpredictable side effects in anyone who would take it.

TAMPER-INDICATING REACTOR SAFING SYSTEM

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Early in 1964, the Hanford Atomic Products Operation of the General Electric Company, which operated the entire Hanford complex at that time, was asked by the United States Atomic Energy Commission to carry out a study for development of an inspection system. The purpose of the system was to assure that production reactors shut down, presumably under international agreement, remained shut down; it was to be applicable to production reactors anywhere in the world.

Constraints of principal importance were that:

1. The inspection system was visualized as including three or four inspections per year by an international inspection team.
2. The system should disclose no more and no less than that the reactor being inspected is not currently being operated and has not operated since the last inspection. (Limited access would be provided to the reactor to the extent necessary for applying the shutdown verification system.)
3. The system should be simple in concept, yet tight enough to be demonstrably secure and to preclude differences of interpretation among lay observers.
4. The system should be unobtrusive as possible to a country's national sovereignty and to its desires to protect classified information; the system should be nondamaging to the reactor.

The requested system was developed in the latter half of 1964, and sufficient laboratory tests were performed with individual components to assure technical feasibility. A systems manual was prepared at the time describing the system and how it would be applied.

In early 1965, the Atomic Energy Commission requested the organization component now known as Douglas United Nuclear, Inc., to carry out a prototypical demonstration in a shutdown reactor of the system developed earlier. The demonstration was to include procurement, training, and field application steps likely to be encountered by an international inspection agency. In other words, the feasibility of the mechanics of the inspection system was to be demonstrated.

The demonstration at Hanford's F Reactor in May 1966 for officials from several interested government agencies completed the basic prototype assignment for determining the operational feasibility of applying the system. The Safing Seal System also had received national and international attention when its development was revealed to the Seventeen-nation Disarmament Conference in Geneva, Switzerland, in April 1966, and the system was described in detail to that organization in August 1966. In November 1966, the installation of the system was demonstrated at Hanford's F Reactor for delegates to the United Nations representing 40 countries. Members of the national and international press attended the November demonstration and recorded the fact that full public disclosure of the system was provided by the government of the United States.

Now for a brief discussion of the concepts and equipment utilized. First, the key to reactor operation is neutrons--lots of them. A neutron-absorbing material such as cobalt, inserted into the reactor core, would be activated in proportion to the amount of plutonium or other weapons material produced. All you really have to know is that the detector, such as cobalt, which was placed in the reactor remained in place between inspections and thus had full opportunity to be exposed to any neutrons present. The problem then becomes one of inventing the "King's Seal."

We decided early on a passive system rather than an active system. In other words, the integrated activation of the neutron-absorbing material, which was included in the wire, and randomly "fingerprinted" components, provide adequate sensitivity and very high reliability. Active concepts, such as ion chambers and accelerometers, with associated circuitry, power supplies, recorders, and possibly telemetry, did not appear to offer notable advantages but would introduce a number of potential disadvantages.

The safing system (see Fig. 1) consists of a safing wire, or tape, attaching hardware at the reactor faces, and a seal. The randomly fabricated "fingerprint" is X-rayed in all cases, and the seal is also photographed. Overlay of before-and-after transparencies provides the check to determine that the assembly has not been removed and repaired, or replaced, between inspections. Neutrons striking the safing tape would be absorbed in sufficient number to induce an easily detectable amount of radioactivity.

Important features of the tape, developed by the then-General Electric Company laboratory component now known as Battelle-Northwest are: 1) the fingerprint pattern of enclosed tungsten or tantalum wires; and 2) a core of lower-melting-point metal. It is fabricated by drawing a tube of copper or a tube of stainless steel over aluminum wire and the "fingerprinting" wire (see Fig. 2). The bimetallic feature inhibits metallic repair

Fig. 1. Typical Installation of Safing Seal System on a Reactor of F Type.

Fig. 2. Safing Tape and X-ray Radiograph.

because of the two melting points. We have found that this feature also inhibits nonmetallic repair. The eddy-current technique, in which a reproducible signal is induced as the safing tape is passed through an electromagnetic coil, provides a backup tool for identification which is especially sensitive to flaws (see Fig. 3).

Fig. 3. Radiograph of Tape Sample before and after Welding, and Eddy-current Traces Made before and after Welding the Sample.

We produced two types of safing tape. One is copper-clad, and the other stainless steel-clad. Both have the aluminum core and the enclosed "fingerprint" pattern. The copper-clad tape is for use where flexibility is desirable, as around external components such as control-rod drives or valve-drive handles. The stainless steel-clad tape is used internally, where

the environment might be corrosive, as could occur with concurrent high gamma activity and moist air, resulting in nitric acid formation.

The specific system which we developed was applied to a double-ended reactor.

Figure 4 shows the hardware components in the "Safing Seal Assembly." This assembly secures the end of the safing tape in the serrated jaws of a clamp (collet), provides a barrier (which itself is finger-printed internally) against access to the tape, and provides a base for the seal which will reveal tampering in case the hardware is disassembled. Also shown is a joint shield--a method for joining safing-tape sections outside the reactor, as for example around control-rod drives or between process channel assemblies. The sample tape and seal components may also be employed on a one-ended reactor with appropriate attaching hardware. For example, one could envision the use of an expanding-mandrel device on a one-ended channel, with an epoxy seal then applied over the bolt head. The seal, incidentally, is formed in the field with a rapidly hardening, clear epoxy, into which are mixed some metallic scraps to form a random pattern for X-ray (see Fig. 5) and photography (see Fig. 6). Polaroid film, available also as transparencies, has been found to be ideal for field use for radiography as well as for photography.

Activation in a production reactor indicating production of only 1/100,000th of a critical mass of plutonium during the exposure interval can be easily detected by use of a portable radiation meter. The system would easily fulfill its function in spite of countermeasures. If of the

Fig. 4. Safing Seal Assembly.

Fig. 5. Making Radiographic Fingerprint of Seal.

Fig. 6. Making Photographic Fingerprint of Seal.

order of a half-dozen geometrically distributed channels are equipped with detectors (see Fig. 7), the shielding required to "poison" all detector locations would itself keep the reactor from operating. Assuming this problem were overcome somehow with distorted enrichment location, sensitive instrumentation would still be able to detect the vast quantities of neutrons necessary to produce even a small amount of plutonium. While countermeasure shielding might reduce sensitivity by an order of 100 to 1,000, this would still leave sensitivity within these instruments in the order of tens of thousands.

Fig. 7. Front Face of F Reactor. Interconnecting tapes are emphasized. All channels are safed on the rear face with an intertie running through the VSR pattern. The actual safing of a Hanford reactor requires no more than five channels. One of the channels in the lower right corner is an extra for the convenience of observers at the demonstration.

An international inspection team is visualized as consisting of four members who would make an original installation in up to five days and a return inspection in about two. The individuals would be selected for versatility, with each able to contribute selected skills such as driver-mechanic, photographer-technician, and engineer-physicist. A fourth member would be one who might fill any position. All members would be

versed in all facets of the equipment and procedures. The team would operate from a home base which would provide a laboratory for photographic reproduction, enlargement, and comparative work, as well as any other work deemed necessary by team members to corroborate verification data obtained in the field. The home base would provide secure files for the necessary records. Working files would accompany team members in the field. The team would be equipped to operate independently from its home base for a period of one week to ten days. A large station wagon is capable of carrying the team and the following equipment: portable generator, small projector screen, viewing box, eddy-current equipment, projector, X-ray machine, two tripods, a small refrigerator or insulated box for film, a file case, individual members' equipment such as cameras and files, and the dozen safing seals and associated hardware necessary for an initial installation, or half that number for subsequent verification inspections. A quantity of safing tape would be included, depending upon the scope of the planned installation.

The team which carried out the prototype system demonstration for the U.N. delegation visiting Hanford in 1966 included a composite of talents which we feel is representative of an international team. The team leader was a test engineer who had been involved in several demonstrations of this type performed earlier at F Reactor. The three technician members had had only brief experience with this system. All four team members gained initial familiarity with the system by reading the 1964 development manual. Their performance and findings demonstrated that the inspection can be performed adequately with teams having only a moderate amount of experience, and with only minor variations from the procedures and equipment specified in the original systems manual.

Thus, we feel that the passive system developed and demonstrated at Hanford fulfills the conditions requested: a highly reliable detector of reactor operation which is simple in concept and unobtrusive with respect to national sovereignty.

These safing tape and seal techniques should be applicable also in the monitoring of operating power reactors--for the tagging of fuel inventories or of reactor components whose inactivity between inspection visits would indicate no change in loading since the last inspection.

PANEL DISCUSSION

DR. KINDERMAN: We now have a panel discussion on Nondestructive Techniques for Assay. The panel consists of Warren McGonnagle, Associate Director, Associated Midwest Universities; Bob Beyster, Research Physicist, General Atomic; Russ Heath, Chief, Physics Section, Idaho Nuclear Corporation; Bob Keepin, Leader of the Pulsed Neutron Research Group, Los Alamos Scientific Laboratory; Norm Rasmussen, Professor, Reactor Engineering at MIT.

The plan of the panel is to present three discussions on gamma spectroscopy and related techniques. The persons discussing gamma spectroscopy are Warren McGonnagle, Russ Heath, and Norm Rasmussen.

V NONDESTRUCTIVE REACTOR FUEL ASSAY

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I. INTRODUCTION

Nondestructive techniques for assay of nuclear fuel are becoming increasingly important from the economic standpoint with the advent of commercial nuclear power and from the security and accountability standpoint because of the worldwide use of fissionable material.

There are two independent problems in the assay (nondestructive) of nuclear fuel. One problem concerns the assay of unirradiated fuel (cold). The other problem concerns the assay of irradiated fuel after a period of exposure in a reactor. In this paper each of these problems will be discussed separately.

A. The Unirradiated (Cold) Fuel

A rapid and accurate nondestructive method of assay for fissionable material content is useful and desirable in the fabrication of nuclear reactor fuel elements for the following reasons:

1. The manufacturing of fuel elements requires many different processes, some of which may cause nonuniform fuel distribution within the fuel element. Mistakes and human errors in the total fuel loading can and have occurred.
2. Foreign materials, contamination, and neutron poisons may be introduced into the fuel element intentionally or inadvertently.
3. Enrichment of fuel is of significance in determining loading and positioning of fuel.
4. If for any reason a fuel plate or a number of fuel assemblies contain more fissionable material than is specified, there is danger of an accident during approach to criticality.
5. The reactor physicist needs to know, for use in his calculations, the total amount and distribution of the fissionable and poison materials in the fuel.
6. The transfer of responsibility for ownership and control of fissionable materials to control by private enterprises poses certain financial, safeguard, and accountability problems.

Each of the above requires measurements to be made without damage to the fabricated fuel element or assembly.

Methods to be considered in the discussion on cold fuel assay include: (1) reactivity changes; (2) fission by thermal neutrons; (3) absorptiometry; and (4) gamma spectroscopy.

B. The Irradiated (Hot) Fuel

The determination of burnup in irradiated fuel is of importance for those interested in (1) fuel research programs, (2) economics, (3) reactor design, and (4) nuclear safeguards.

One of the fundamental quantities required in a fuel research program is the burnup of the fuel test specimens. This quantity is required to correlate the change that takes place in fuel with temperature variations, integrated neutron flux, position in the reactor, and other parameters.

The reactor design data are of prime importance in establishing the validity of reactor constants used. This is an integral part of the core design, reactor performance, and core life, which, in turn, is related to the economics of reactor operation. An error in the overprediction of the reactor core life could possibly result in severe economic loss to the reactor designer and manufacturer.

One of the important functions of the International Atomic Energy Agency (IAEA, Vienna, Austria) is to oversee and administer an international safeguards system. This function is carried out through an international system of control and inspection designed to ensure that nuclear materials, facilities, and technology furnished for peaceful uses are not diverted to military applications. From the viewpoint of the IAEA, it is most desirable that no special information about the irradiated nuclear fuel specimen, as far as irradiation history or cooling period is concerned, should be required.

Methods to be discussed for irradiated fuel assay include:

1. activation of foils;
2. spectrometry using
 - (a) the Compton magnetic spectrometer;
 - (b) bent-crystal diffraction spectrometer;
 - (c) Compton coincidence spectrometer;
 - (d) scintillation detectors;
 - (e) solid-state detectors;

3. calorimetry and thermal conductivity;
4. measurement of prompt and delayed neutrons;
5. activation analysis;
6. capture gamma-ray measurements;
7. neutron transmission;
8. use of stable isotopes.

II. COLD FUEL ASSAY

A. Neutron Fission

Forbes and co-workers¹⁻³ developed a method of nondestructive fuel assay using neutrons from the thermal column of the Materials Test Reactor (MTR). The method is based primarily on the fact that the fission rate in U^{235} depends on the amount of fissionable material in the specimen and the thermal-neutron intensity. The fuel plate or fuel element to be assayed was placed in the beam of thermal neutrons and the number of fast neutrons produced in the fission process determined. Determinations of the fuel content were made by comparing the fission of a specimen with that of a standard specimen of known content of fissionable material. It is also possible to have neutron absorption by nonfissionable materials (poison) that effectively reduce the thermal-neutron beam intensity to which the fissionable material is exposed. Therefore, an additional measurement of the attenuation of the thermal-neutron beam as it passes through the fuel element is required. The attenuation of the thermal neutrons is a measure of the total cross section of the fuel plus the poison. A detector in the unattenuated beam provided a monitor of the beam strength for both fission rate and transmission data.

The absorption of neutrons follows an exponential law given by

$$I = I_0 e^{-\Sigma x} = I_0 e^{-(\Sigma\mu + \Sigma\rho)},$$

where

I = intensity of the transmitted beam;

I_0 = intensity of the incident beam;

x = absorber thickness;

Σ = macroscopic cross section;

$\Sigma\mu$ = total cross section of the fissionable material per unit area normal to the incident beam;

$\Sigma\rho$ = total cross section of poison per unit area normal to the incident beam.

The nuclear fuel itself causes some self-shielding. Thus, the attenuation of the thermal beam transmitted through the specimen reduces the fission rate in that portion of the fuel located farthest from the incident beam. The response of a detector to a given amount of fissionable material will be greatest for the fissionable material closest to the beam. Forbes¹ gives the mathematical treatment.

A boron trifluoride counter was used to monitor the thermal-neutron beam from the reactor as well as to measure the transmission through the fuel. Fission neutrons were detected by two boron trifluoride counter assemblies placed on opposite sides of the fuel assembly and in a plane 90° to the thermal beam. These detecting assemblies consisted of two wooden boxes, 6 x 12 x 12 in., filled with paraffin and lined internally with a 20-mil cadmium sheet. The boron trifluoride counters were placed in the center of the paraffin. The cadmium sheet absorbed the thermal neutrons striking the box. The fission neutrons after passing through the cadmium were subsequently moderated by the paraffin to thermal or near-thermal energy and detected. The fission-neutron counter and a transmission counter were automatically normalized to a fixed number of neutrons incident on the fuel assembly.

Fast-neutron absorption in the side plates of the fuel element was negligible. The errors resulting from the various causes in general were less than one percent. Forbes used a three-point spot check along the length of the element, plus a determination of the effective length of the fuel plate. The length measurement was a source of some error.

This test is no longer in use at MTR because: 1) tests could only be made when the reactor was operating at a steady power, 2) the thermal column came out of the top of the reactor where space was limited, and 3) the equipment had to be moved into position each time a test was made.

B. Reactivity Measurements

The term "danger coefficient" originated in the early days of the Manhattan Project. It was recognized at that time that there was the possibility of materials intended for reactor use containing impurities of high neutron-absorption cross section which could not be detected by conventional chemical means. It was considered necessary to measure the neutron-absorbing properties of reactor materials.

In the danger-coefficient method a low-power reactor is used. The reactor is operated near criticality and the reactivity determined from the reactor period. The material under investigation is inserted in the reactor and the new reactor period determined. By comparison with a standard, the change in reactivity due to a given material can be determined.

Three techniques are in general use for measuring reactivity changes in nuclear reactors: the pile-period technique, the pile-oscillator technique, and the automatic-flux-control technique. The basic theory of such measurements can be found in any textbook on nuclear reactor theory.⁴⁻⁶

The pile-period technique was historically the first used to determine changes in reactivity. A pile is brought to criticality, then a perturbation is introduced. The reactivity is computed from the rate of increase of the neutron flux, using the kinetics equations developed in reactor theory. Any extraneous neutron source will introduce errors into the measurements.⁷ The time required for each measurement is long, as it is necessary to wait for all transients to die out after introducing the change.

In the pile-oscillator technique, a specimen whose reactivity effect is to be measured is alternately inserted and retracted from the core region of the reactor, either sinusoidally or stepwise. The change in flux amplitude and phase is recorded. These quantities are functions of the nuclear properties of the specimen. By comparison with a standard specimen, the cross section of the unknown specimen is found. This technique has the advantage of reducing the effect of any long-term drift in the power level of the reactor. If the flux level of the reactor is increasing linearly, the effect of the increase may be eliminated by integrating the data over the proper interval.

The third technique used is that of automatic flux control. In this technique the flux level of the reactor is kept constant by means of a servo system that regulates a lightly absorbing control rod. Any change in the reactivity of the pile will result in a change in the position of the rod. The magnitude and direction of this position change will be a measure of the magnitude and sign of the reactivity perturbation. As with the oscillator technique, the cross sections are obtained by comparison with a standard specimen. The precision of reactor measurements is extremely high. The theoretical limit to the precision of reactivity measurements is determined by the statistical variation of the neutron population within the reactor core.⁸

It has been shown that the error is one milligram for each kilogram of fuel material contained in the reactor. However, as this is a comparative test, any error in the standard specimen will affect the accuracy of the measurement. This technique cannot distinguish between the various types of fissionable material: Pu^{239} , U^{235} , or U^{233} . Temperature variations, barometric-pressure changes, and the self shielding can affect the accuracy of the measurements.

Fast and co-workers⁹ at the Phillips Petroleum Company, Atomic Energy Division, have used a critical assembly, the Reactivity Measurement Facility (RMF), to evaluate thorium slugs for U^{233} content and ETR fuel elements for U^{235} and boron. The RMF¹⁰⁻¹² is a critical test facility

located in the Materials Test Reactor (MTR) canal. Gustavson and Sherwin of Aerojet General Nucleonics¹³ have used the AGN-201 reactor to evaluate properties of reactor-fuel materials and moderators nondestructively. Gustavson and Sherwin give the following as typical results: (a) differences of less than 0.005% in uranium enrichment; (b) differences in uranium impurity levels equivalent to less than 0.3 ppm; and (c) differences in beryllium impurity levels equivalent to less than 0.2 ppm boron or less than 300 ppm iron.

Large fuel elements are more difficult to evaluate accurately than small elements. This is because a change in concentration will cause a greater or lesser reactivity change depending on the physical location of the change within the fuel element. The existence of several nuclides in a large test element presents a problem which in practice is insoluble if it is desired to determine changes in all of the nuclides.

For most large fuel elements, the radial distances of the fuel element from the reactor centerline changes the reactor sensitivity to variations in poison or fuel. Also, changes in poison or fuel concentrations that take place in discrete positions axially can cause error because the sensitivity is a function of axial position. If a change in concentration occurs in only a small volume in the test element, there may be strong "self-shielding" effects that usually cause the sensitivity to be less than the sensitivity to a uniform change.

In the design of a fuel-assay reactor, the following should be considered: 1) maximum sensitivity of reactivity to changes in the central test region, 2) ease of handling and evaluating of large fuel elements, 3) ease of evaluation of different elements that cause large changes in the net reactivity of the reactor, and 4) adequate and safe means of control without distorting flux distributions in the inner regions of the reactor.

From this discussion it is evident that relative changes in U^{235} content as low as a few tenths of 1% can be determined if errors in reactivity can be as low as 10^{-5} and if the relative changes in poison content are reasonably small.

C. Absorptiometry

X-ray and gamma-ray absorptiometry provides a simple, rapid, and fairly accurate method for the nondestructive analysis of reactor fuel materials.

When monoenergetic X or gamma rays pass through material, the intensity decreases. The decrease in intensity is given by Lambert's law:

$$I = I_0 e^{-\sum \mu_i M_i},$$

where

I = intensity of transmitted radiation;

I_0 = intensity of incident radiation;

M_i = mass per unit area of each material in the absorber;

μ_i = mass absorption coefficient for each material.

The value of the constant depends on the material and the energy of the X- or gamma-ray photons.¹⁴

X rays and gamma rays can be used to detect one element in the presence of another of different atomic number. This differential absorption is due to the fact that the absorption coefficient for each material is different for a given energy. Generally stated, to preserve sensitivity it is best to use the softest radiation that has the required penetration.

It can be shown that for variation in chemical composition, such as in the case of metal alloys,

$$\frac{I_2 - I_1}{I_2} = t_2(\mu_2 - \mu_1),$$

where

I_1 = intensity transmitted by one material;

I_2 = intensity transmitted by the second material;

μ_1, μ_2 = respective linear absorption coefficients;

t_2 = thickness of material 2.

By making the difference in the linear absorption coefficients large, a high sensitivity can be achieved. This is done by choosing the energy of the penetrating radiation which makes the value of $\mu_2 - \mu_1$ a maximum.

With the continuous radiation emitted by an X-ray tube, an appreciable difference in atomic number is necessary between two materials to give sufficiently large differences of the linear absorption coefficient to obtain good contrast--for example, of uranium in aluminum. The contrast can be enhanced if monochromatic radiation is used.

Absorptiometry has been applied to fuel-loading and fuel-distribution measurements. A prototype system similar to a system developed at Harwell was evaluated for measuring the UO_2 distribution in UO_2 -stainless steel matrix plates. This work was done on fuel elements for the Argonne Advanced Research Reactor being designed at Argonne National Laboratory.

The system is shown in Fig. 1. An Am^{241} source with a gamma-ray energy of 59 keV and a Co^{57} source with a gamma-ray energy of 122 keV were used. The 59-keV source gave a greater sensitivity to UO_2 concentration, as expected from the theoretical discussion. However, the lower energy showed a greater sensitivity to possible changes of cladding thickness. The 122-keV source was relatively insensitive to changes of cladding thickness and still gave adequate sensitivity to UO_2 concentration. Plate loading variations of $\pm 3\%$ over a 0.5-cm^2 area could be detected. A facsimile recording made with the system is shown in Fig. 2.

Fig. 1. Scanning and Recording System Using Quantizer.

Fig. 2. Facsimile Recordings (above) and Autoradiographs (below) of Fuel Plate.

A continuous-scanning, X-ray attenuation system has been developed by Oak Ridge National Laboratory, in which an X-ray machine is used as the source. With the high intensity available, the system is able to evaluate areas as small as 0.007 in.² at scanning speeds up to 150 in./min. Oak Ridge has used their scanning system primarily on aluminum-U₃O₈-type fuels.

D. Natural Gamma Activity

The natural gamma activity of reactor fuels has been used extensively for fuel assay by a number of laboratories.^{15,16} In a sample of highly enriched uranium containing 93% U²³⁵, 5.5% U²³⁸, 1% U²³⁴, and 0.5% U²³⁶, the U²³⁵ isotope contributes only 3% of

the total disintegrations. But, examination of the gamma-ray spectra of these uranium isotopes shows that the U²³⁵ isotope is the primary source of gamma radiation. The gamma-ray spectrum of highly enriched uranium obtained with a NaI detector is shown in Fig. 3. Figure 4 shows the gamma-ray spectrum of highly enriched uranium obtained with a Ge-Li detector. The 184-keV gamma ray is entirely due to U²³⁵ and is especially useful for U²³⁵ assay. In normal uranium the U²³⁵ isotope produces approximately 2% of the disintegrations, and its presence is still detectable in the

gamma-ray spectrum. The intensity of the 184-keV gamma ray is directly proportional to the U²³⁵ content, which can therefore be measured by gamma-counting techniques.

Fig. 3. Spectrum of U²³⁵ Obtained with a NaI Detector.

Fig. 4. Spectrum of U²³⁵ Obtained with a Ge-Li Detector.

Figure 5 illustrates a technique¹⁷ for counting fuel plates in which uranium foils are used as standards. The difference in self-absorption is measured and corrected for by use of the equation

$$\frac{C_t}{C_0} = \frac{1 - e^{-\mu t}}{t}$$

When the self-absorption correction is applied, there is a nearly linear relationship between U^{235} content and the counting rate.

Fig. 5. Technique for Determining U^{235} Content.

The block diagram of a scanning system for counting the natural gamma radiation from reactor fuels used by the Quality Evaluation Group at Argonne National Laboratory is given in reference 18. Figure 6 shows the equipment used for scanning plate-type fuel elements. Some typical results obtained with plate-type elements, uranium foils being used as standards, are shown in Table I. The U^{235} content was computed from the equation

$$\text{Unknown} = (U_{\text{std}}) \frac{(C_{\text{O plate}})}{(C_{\text{O std}})}$$

A computer program for calculating the U^{235} content and the statistical error has been developed for use with the scanning system.

Fig. 6. Scanning of Fuel Plate.

TABLE I. Comparative Results

Plate Number	U ²³⁵ Content (g)		
	Gamma Counting	Chemical and Mass Spec. Analysis	Fabrication Data
AD-9	17.01 ± 0.08	16.953	17.01
AP-11	17.74 ± 0.12	17.735	16.68
605	15.10 ± 0.30	15.07 ± 0.08	
598	18.43 ± 0.37	18.17 ± 0.11	
500 C	4.24 ± 0.08	4.10 ± 0.02	

III. HOT FUEL ASSAY

A. Activation

In the activation technique, a uranium or plutonium foil is attached on the surface of a fuel element. The foil is removed after discharge of the element from the reactor. The quantities of U²³⁵ burned out and plutonium produced are determined by the analysis of the alpha spectrum of the foil. Because of low activity of U²³⁵, the foil must be enriched in the U²³⁵

isotope. In cases of enriched fuel produced by the diffusion process, U^{234} is also present, and the background of the U^{234} peak has a disturbing effect on the resolution of both U^{235} and U^{238} .

Some difficulties with the technique are that: the uranium and plutonium must be clad; the resolution of the alpha spectrum is low; enriched uranium must be used to obtain sufficient alphas; U^{236} is produced; the high beta-background exerts an influence on the alpha spectra, and there is self-absorption.

Monitors of cobalt alloy wire have been used to determine burnup.

Another method of assaying irradiated fuel is to measure the intensity of one of the several gamma lines from a fission product of known

TABLE II. Resolution of Compton
Magnetic Spectrometer

Gamma-ray Energy (MeV)	Half-width at Half-maximum (%)
0.48	4.15
0.78	2.0
1.14	1.45
2.74	1.15

fission yield. Owing to the extremely complicated spectrum, a high-resolution instrument is needed for this analysis. Groshev and Demidov^{19,20} have used a magnetic Compton spectrometer for such an investigation. The order of magnitude of the resolution of such an instrument is as shown in Table II.

In the Compton magnetic spectrometer the gamma rays from the irradiated material are converted to electrons by Compton scattering in a special converter foil placed in the spectrometer. The spectrometer is designed to measure the momentum and distribution of electrons ejected from the foil into a prescribed solid angle by the incident photon beam. The collimated photon beam impinges on a foil (such as beryllium) positioned in the gap between the pole pieces of an electromagnet. Electrons and positrons are ejected from the foil with an energy and angular distribution which depends on the incident photon energies, the foil thickness, and the atomic number of the foil. For a given magnetic field in the gap between pole faces, electrons of a particular momentum range are focused at the exit slit. However, because of the interposition of the defining baffle, only a small fraction of these electrons, namely, the electrons contained in a small solid angle defined by the baffle opening, reach the detector.

Such an instrument permits the evaluation of photon energies in a range extending from approximately 0.2 to 12 MeV with an accuracy of 2%. Also, the photon intensity for a specific energy can be measured to an accuracy of 10%. The price paid for higher resolution is a decrease in efficiency. For example, in a typical magnetic spectrometer a monoenergetic photon flux of the order of 100 photons/cm² will produce approximately one detectable event. Film, solid-state devices, and scintillators have been used as detectors.

Another way of analyzing the spectrum is with a bent-crystal diffraction spectrometer. The Dumont type has very good resolution, but requires extremely strong sources, and the construction is not simple. The total efficiency of a diffraction spectrometer is inherently very low (10^{-8} to 10^{-10}), the efficiency decreasing with increasing energy.

Rasmussen and Cohen²¹ used a 6-m bent-crystal spectrometer. The uranium ($K_{\alpha} - 1$) X ray has an energy of 98.442 keV and the plutonium ($K_{\alpha} - 1$) X ray has an energy of 103.7 keV. Line intensities were measured with a sodium iodide scintillation detector. The two K_{α} radiations previously mentioned were separated by 1.6 cm on the spectrograph focal circle. The background radiation was discriminated by using a relatively small sodium iodide crystal (2 cm in diameter by 2 mm thick) and a single-channel pulse-height analyzer following the detector.

Johnson and Vrablik²² and Kristiansen and Rogeberg²³ have used a crystal spectrometer to evaluate burnup. Their results indicate that the burnup must be rather high to detect the presence of plutonium and the cooling time must be relatively short.

In the Compton-recoil spectrometer a well-collimated beam of gamma rays impinges on a detector (scintillator crystal or solid state), and the backscatter quanta are detected by a second detector. A coincidence circuit is used to select and count Compton events of definite backscatter angle. By selecting only Compton events backscattered at an angle near 180° , a single peak for monoenergetic gamma rays is obtained. Selection of suitable events is done by accepting only those pulses from the second detector which are in coincidence with the back-scattered quantum detected in the first detector. This spectrometer is characterized by its efficiency, its response function, and its energy resolution. The total intrinsic efficiency is approximately 10^{-3} at maximum, decreasing toward higher and lower energies. This is due to absorption losses--especially for backscattered quanta at lower energies--and to the decreasing Compton cross section at higher energies.

The Compton-recoil spectrometer has an efficiency comparable to the Compton magnetic spectrometer (0.1% per incident photon) but considerably better resolution.

Gamma scanning for various fission products has been used or proposed for measuring burnup. Frequently, Ce-Pr-144, with a half-life of 284 d and 90-keV gamma line, is used for burnup determinations, because the self-absorption in the fuel element is lower at the higher energies. Furthermore, cerium has no tendency to migrate to the surface in the high temperature fuels.

Ca-Ba-137 with a half-life of 30 yr is very advantageous for use in burnup analysis because of its long half-life and approximately equal yields

for U^{235} and Pu^{239} fission. For normal fuel-element-utilization time of up to three years, the time factor can be estimated with good accuracy without information on reactor operation. Because of the quality of fission yields for the mass number 137, the determination of Cs^{137} leads directly to the sum of U^{235} and the Pu^{239} fissions in the fuel containing only these fissionable isotopes. For practical cooling times of the order of weeks, the 661-keV line is very difficult to measure, due to the neighboring much stronger gamma lines from other fission products. Also, there is the possible influence of the migration of Cs^{137} in the fuel.

Westinghouse has used gamma scanning to determine burnup in a Yankee-type reactor.²⁴ Their results may be summarized as follows: "Within certain limitations gamma scanning measurements can be used to determine relative burn-up distribution, local peaking factors, the maximum-to-average burn-up ratio, and gross core asymmetries." In the Yankee-type core, a simple one-to-one correlation between gamma activity and burnup does not exist.

By use of the burnup and gamma activity of a standard irradiated assembly as a reference, and the assumption of a one-to-one correlation between burnup and gamma activity, it was determined that the gross gamma scans made in the assembly center-holes predicted the assembly average burnup to within approximately 8% on the average. The gamma scans under-predicted burnup by as much as 17% for low-burnup assemblies, and over-predicted burnup by as much as 18% for high-burnup assemblies. These discrepancies, which include gross errors caused by the nonuniform cobalt distribution, can be reduced with the use of a burnup-to-gamma correlation. For Yankee Core 1, application of the burnup-to-gamma correlation to the assembly-average gamma activities reduced the average discrepancy from 8 to 4%, and narrowed the range of the discrepancies from -12 to +18%, down to -5 to +10%. If the gamma scans were corrected for cobalt non-uniformity, the difference between the burnup and the gamma distribution would be somewhat smaller than the values given above. Although the assembly-average gamma activities were inferred from volume-weighted pellet burnups measured in the central and corner subassemblies, the agreement between the burnup and gamma-activity distributions is quite good.

The use of gamma scans overpredicts the maximum point-to-average burnup ratio by 14% if the gamma activity is taken to be directly proportional to burnup. Again, the use of the burnup-to-gamma correlation given previously sharply reduced the observed discrepancy. Gamma scanning measurements of fuel rods do not reliably predict gross burnup asymmetries in the xy-plane unless the measurements are free from the bias which can be introduced by nonuniformities in the fuel-rod cladding.

Owen²⁵ has used the gamma activity of two specific fission products for measuring burnup. A fission product of short half-life gives a measure of the power distribution in the fuel for the period immediately before discharge, while a product with long half-life gives a measure of the power integrated over the whole irradiation period. During irradiation of fuels in a reactor, the fission products of short half-life build up to a state of secular equilibrium or saturation after a few half-lives. The power reactor changes their concentrations to new values characteristic of the new power level after a few half-lives. A buildup of fission products of long half-life is approximately linear for about one half-life. Owen has chosen two fission products which have the following properties: characteristic gamma radiation which can be readily discriminated from the general background, and high percentage yields from the main fissile isotopes.

B. Activation Analysis

To evaluate the feasibility of fission product activation analysis for determinations of U^{235} burnup in fuel, an extensive examination of all the stable fission products and their stable daughters was made. The Rh^{103} isotope was chosen because of its high thermal-neutron activation cross section (140 b), its short half-life (44 sec), and the relatively high fission yield (3%) of its parent Ru^{103} , which has a 39.7-day half-life. A detailed analysis of this technique and calculations can be found in reference 26.

This technique has the following limitations: (1) a waiting time of about six months after shutdown is required so that essentially all Ru^{103} has decayed to Rh^{103} ; (2) only the analysis of highly enriched fuel plates will produce any meaningful data because of the extremely high standard counting deviation for low-enrichment fuel plates; (3) conventional counting techniques are inefficient for counting the 556-keV gammas. For 90% enriched fuel plates, the resulting statistical error is significant. A 10.7% standard deviation in counting was calculated for the case of 90% enrichment and 2% burnup, using a solid-state detector and 2π geometry. Scintillation detectors are not practical in this case because of poor resolution and high gamma background from the irradiated fuel plate under test.

C. Prompt and Delayed Neutrons

In the fission process both prompt and delayed neutrons are emitted. The measurement of prompt and delayed fission neutrons had been proposed as a method for measuring burnup. The basic principle is the measurement of the fast fission neutron emitted as a result of thermal-neutron irradiation of the irradiated fuel elements. Such a measurement is sensitive only to the presence of unknown materials. Although the method will measure directly the total fissionable material in the specimen, one cannot directly determine the U^{235} and plutonium contents without other knowledge.

A refinement of the above technique would be to measure both the total fission neutrons and the delayed neutrons (see Table III). The fast-neutron yields from Pu^{239} and U^{235} will be markedly different in these two measurements. The number of fast neutrons emitted per thermal fission are 2.43 and 2.89 for U^{235} and Pu^{239} , respectively. The number of delayed neutrons per fission are 0.158 and 0.0061 for U^{235} and Pu^{239} , respectively.

TABLE III. Fission Yields of Ba-La and Ce-Pr

Fissile Material	Type of Fission	Yield of Ba-La Chain (%)	Yield of Ce-Pr Chain (%)
U^{235}	Slow	6.44	6.0
U^{235}	Fast	4.58	5.0
U^{238}	Fast	5.7	4.9
Pu^{239}	Slow	5.68	5.28
Pu^{239}	Slow	5.0	

D. Capture Gamma Rays

When a thermal neutron is captured by a nucleus, the resultant compound nucleus is left in an excited state from which decay occurs promptly with the emission of capture gamma rays. Capture gamma-ray spectra are complex, containing both low- and high-energy gamma rays. In the case of fissionable materials, the prompt gamma spectrum consists of two parts: the prompt gamma rays emitted in the fission process, and the capture gamma rays emitted in the neutron-gamma reaction. The prompt gamma spectra resulting from the fission reaction in U^{235} and Pu^{239} are essentially identical. However, the capture gamma-ray spectra from U^{235} and Pu^{239} in the neutron-gamma reaction are different.

The basic principle of the method is to irradiate the fuel elements with thermal neutrons and detect the capture gamma-ray lines characteristic of U^{235} and Pu^{239} . One of the problems in this technique is that the capture gamma-ray intensity from the fuel element must not be much weaker than the fission product activity of the fuel element. The sodium iodide scintillators cannot resolve the individual capture gamma-ray lines, but use of solid-state detectors, which have greater resolution, may be used.

E. Neutron Transmission

The attenuation of neutrons in spent fuel elements occurs due to attenuation by U^{235} , Pu^{239} , and the fission products. However, the main effect is due to the combined effects of U^{235} and Pu^{239} . Therefore, measurement of the thermal-neutron transmission through the spent fuel

element can provide a measurement of the fissionable material present. Practical measurements of thermal-neutron transmission should be possible using an external neutron beam from a reactor, or thermalized neutrons from a neutron generator, or one of the stronger isotopic sources. In this method it is necessary to know the integrated neutron dose or burnup to which the specimen has been subjected. In many reactors, neutron flux varies along the axis. In addition, local variations due to self-shielding and interference from other irradiation specimen, lead to a variation in the burnup along the axis of the specimen.

F. Stable Isotopes

There are a number of problems in using radioactive isotopes for burnup analysis. One of the major problems is the influence of reactor operation and cooling time on the test results. The use of stable isotopes for burnup analysis has been proposed. In the use of stable isotopes a detailed irradiation history is of secondary importance and cooling time is unimportant. Also, there are more precise nuclear data for the stable fission product nuclei. Stable isotopes are being used in conjunction with mass spectrometry for destructive burnup analysis.

There are many factors which dictate the choice of a stable isotope for use as a measure of burnup. The main considerations are concerned with the nuclear properties of the isotope, and it is essential that the number of atoms of the nuclide formed in the specimen is directly related to the number of fissions that have occurred during irradiation.

Two elements with isotopes which meet most of the above requirements are molybdenum and neodymium. Neodymium is unlikely to undergo any migration under irradiation, and the precursors that do exist are of short half-life and of low neutron capture cross section.

Although there are about 30 stable fission products which have known fission yields, the possible choice of isotope(s) is limited to molybdenum and neodymium. The neutron cross sections of most of the molybdenum isotopes are significantly lower than those in neodymium.

The isotopes of molybdenum (mass numbers 97, 98, and 100 are formed in nuclear fuel by fission) have the advantage of high fission yields, low cross section for neutron capture reaction, no false precursors, and both Mo^{98} and Mo^{100} are primary fission products.

The determination of the amount of a given stable isotope might be determined by such nuclear techniques as activation analysis or capture gamma-ray analysis. However, much experimental work must be done before these methods would become feasible or useful for the nondestructive determination of burnup.

G. Slowing-down Spectrometer

Chandramoleswar and co-workers²⁷ have used the slowing-down spectrometer for assay of Pu^{239} . In the energy range from 0.01 to 1.0 eV, the fission cross section of U^{235} and Pu^{239} are similar, except at 0.3 eV, where Pu^{239} has a resonance whose value is about twenty times that of U^{235} . This difference can be used to determine the Pu^{239} content of irradiated fuel if the relative fission rates in Pu^{239} and U^{235} are measured as a function of the neutron energy.

Chandramoleswar used a Van de Graaff accelerator as a source of pulsed neutrons. Short bursts of fast neutrons were injected periodically into thick lead. Neutrons of various energies, having a small energy spread about a mean energy, can be obtained as a function of time from each burst. The experimental results indicate that it is possible to detect plutonium in an irradiated specimen. However, the system has not been optimized as yet, so that the sensitivity and resolution of the method is not known as yet.

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THE POTENTIAL OF HIGH-RESOLUTION GAMMA-RAY SPECTROMETRY FOR THE ASSAY OF IRRADIATED REACTOR FUEL

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The purpose of this presentation is to provide a brief summary of the state-of-the-art in the development, use, and possible application of the lithium-ion drifted germanium gamma-ray detector to the problems in the area of fuel assay. In any consideration of potential nondestructive techniques which might be applied to the assay of nuclear fuel, the measurement of the concentration of fission product nuclides by means of techniques of gamma-ray energy spectroscopy represents one of the most sophisticated techniques at our disposal. It not only is in a highly developed state but is relatively simple in concept.

The developments of the lithium-ion drifted detector and of low-noise cryogenic electronics during the past year or so for applications in basic physics studies have resulted in improved energy resolution which makes it possible to examine gamma-ray spectra in much greater detail. This increase in resolving power is of particular importance in the analysis of the gamma-ray spectra of gross fission products because of the complexity resulting from the presence of many radioactive isotopes in such a sample.

Prior to any attempt to describe how this capability may apply to the problems under discussion today, let us briefly consider the basic techniques to be employed. Figure 1 is a simple diagrammatic representation of a lithium-ion drifted germanium gamma-ray detector. A P-I-N structure is created by drifting lithium ions through a single crystal of germanium or silicon material. The most important property of this structure is that

Fig. 1

Diagram Showing Structure of P-I-N Junction Detector. Gamma rays entering the intrinsic region, upon interaction, transfer part or all of their energy to an energetic electron. The electron energy is subsequently transferred to electron-hole pairs which are collected at the electrodes of the device. This process creates a current pulse containing a quantity of charge which is proportional to the energy of the gamma ray detected in the sensitive volume of the detector.

it acts as a solid-state ionization chamber. If a gamma-ray photon enters the intrinsic region of the device and interacts with an electron by the photoelectric, Compton, or pair-production process, some or all of the gamma-ray energy will be imparted to the electron. Subsequently this energy will be expended in the production of electron-hole pairs. If an electric potential is applied to the device, the generated charge will be collected at the electrodes, resulting in a current pulse. The amount of charge collected will be proportional to the energy left in the sensitive volume and hence to the energy of the gamma ray detected. By amplifying this minute amount of charge with low-noise electronic pulse amplifiers and measuring the amplitude distribution of pulses resulting from the detection of gamma rays, one may obtain a measure of the energy spectrum of gamma rays emitted by a radioactive source. The techniques of gamma-ray energy spectroscopy using pulse amplitude techniques have been well developed in the application of the scintillation detector.

To illustrate the results which can presently be obtained with such spectrometers using the best available electronics, Fig. 2 shows a 4096-channel pulse-height distribution representing the gamma-ray energy spectrum of the radiation emitted by gross fission products. To obtain these data a sample of U^{235} was irradiated for 30 min in the MTR and the gamma-ray spectrum obtained at one hour after removal from the reactor. The energy range represented is from about 20 keV to 3 MeV. A detailed analysis of this pulse-height spectrum indicates the presence of over 150 individual peaks representing gamma rays emitted in the decay of 40 different radioactive fission product isotopes. Using a knowledge of the decay characteristics of each nuclide, gamma-ray energies and intensities are used to identify individual isotopes. The key in the upper right-hand corner of the figure lists the 35 most intense isotopes; the individual gamma-ray peaks characteristic of each isotope are indicated alphabetically on the ascending energy scale. The primary purpose for using the short-lived fission product gamma-ray spectrum was to illustrate the complexity of fission products as a source of radiation and the extreme detail which can be obtained with a high-resolution gamma-ray spectrometer. It is of interest to note that the increased precision afforded by the Ge(Li) spectrometer has made it necessary to remeasure the gamma-ray spectrum of most nuclides to obtain more precise values for the intensities and energies of gamma rays emitted in the decay of each fission product nuclide. This example should also serve to point out the large volumes of data that must be handled in the analysis of such results. The successful application of these techniques on a large scale for the solution of applied problems will require automated data-processing techniques.

At the present time considerable attention is being given to the development of specialized detector systems to improve the performance of Ge(Li)

spectrometers for the analysis of complex gamma-ray spectra. Figure 3 is a detailed pictorial of a gamma-ray spectrometer employing a large sodium iodide scintillation detector as an annulus for the rejection of Compton electrons in a gamma-ray spectrum. As shown in the lower left-hand corner of the figure, the arrangement employs a germanium detector which is surrounded by an annulus of sodium iodide. The gamma rays are collimated into the central Ge(Li) spectrometer. In this configuration, if a Compton event occurs in the central detector, the scattered Compton photon will have a reasonable probability of being detected in the large NaI detector surrounding the Ge(Li) spectrometer. If the two detectors are operated in coincidence, such events can be identified and rejected in the final analysis by the pulse-height analyzer.

The result of this method of operation is shown in Fig. 4. In this figure, pulse-height spectra resulting from the measurement of Co^{60} gamma rays, obtained with a Ge(Li) spectrometer with and without a NaI(Tl) annulus, are shown for comparison. Examination of these two curves indicates the nature of the result of the rejection of Compton events by the NaI annulus detector. The two well-defined peaks result from events which leave the total energy of the gamma ray in the central detector. The continuous distribution of pulses down to zero energy results from Compton events and represents information which only serves to complicate the shape of the observed pulse-height distribution. If this continuous distribution of pulses could be eliminated, our ability to recover information from a complex spectrum would be greatly enhanced. It is our opinion that this principle could be refined to produce a further reduction in the Compton continuum by a factor of ten. This would considerably improve our ability to analyze complex spectra.

To illustrate the importance of this technique to the analysis of gross fission product spectra let us observe the spectra shown in Fig. 5. In this figure we have plotted the gross fission product gamma-ray spectrum resulting from an 8-hr irradiation of U^{235} and a decay of 17 days following removal from the reactor. The two spectra compare results obtained with and without the use of a Compton rejection mantle. Close inspection of these two spectra shows that considerable additional detail is apparent in the lower curve. To illustrate the enhancement, the lower curve is labeled only with those peaks which were not detectable without the Compton rejection mantle.

Since the techniques which have been described above can be used to assay irradiated fuel for the content of specific fission product nuclides, we must ask ourselves what significance can be attached to this class of information. Without a detailed treatment of the various factors which relate fuel history to fission product distribution, let us first examine some typical fission product spectra obtained from irradiated samples of U^{235} and Pu^{239} as examples of the experimental problem.

Fig. 3. Gamma-ray Spectrometer System Employing a Lithium-ion Drifted Ge Detector Surrounded with an Annulus of Sodium Iodide. A collimated beam of gamma rays enters the central detector. Scattered gamma rays resulting from Compton processes in the central detector are rejected by coincidence circuitry if the scattered photon is detected in the NaI annulus detector.

Fig. 4. Pulse-height Spectrum of Radiation Emitted by a Co^{60} Source. The two pulse-height spectra illustrate the effect upon the shape of the spectrum produced by the Compton suppression technique.

Fig. 5. Pulse-height Spectrum Representing the Gamma-ray Spectrum of Gross Fission Products with and without the Use of a Compton Suppression Annulus Detector System. The peaks identified in the lower curve are those which could not be identified in the spectrum without suppression of the Compton continuum with an annulus detector surrounding the Ge(Li) spectrometer.

Figures 6, 7, and 8 show gamma-ray spectra obtained from three samples of irradiated material, respectively the spectrum obtained from a sample of U^{235} irradiated to 5% burnup at a flux of 2×10^{13} n/cm²/sec, with a cooling time of 180 days; the spectrum from a sample of U^{235} irradiated to 50% burnup at the same flux with a cooling time of 250 days; the spectrum from a sample of Pu^{239} irradiated in a flux of 2×10^{13} n/cm²/sec with a cooling time of 180 days. Examination of the spectrum in Fig. 6 shows that little structure is evident above the Nb^{95} line at 765 keV. Comparing this region with the U^{235} data representing 50% burnup, we see additional detail in the energy region above the Nb^{95} line. This indicates that, as might be expected, the relative concentrations of different nuclides are dependent on nvt. Observation of the Pu^{239} spectrum in Fig. 8 shows that the region of interest becomes considerably more complex in nature. If the two high-burnup spectra of Figs. 7 and 8 are compared, we see that, for comparable nvt, the concentrations of particular nuclides are clearly different for Pu^{239} and U^{235} . In particular, the nuclides Ru^{106} , Ag^{110m} , and Eu^{154} appear to be greatly enhanced in the fission product spectrum of plutonium. This results from differences in the mass-yield curve for the two fissile nuclides.

The relative abundances of individual fission product nuclides at a particular time will be a function of a number of related variables. These include the average flux in the reactor environment, total nvt, the energy spectrum of the reactor, the mass-chain yield for the nuclide in question, the magnitude of the capture cross section of the nuclide, and the time after removal from the reactor. As an example of one of these factors let us examine Fig. 9. In this figure we have plotted the ratio of the mass-chain yield for thermal fission of Pu^{239} to U^{235} as a function of mass number. An examination of these data would lead us to predict that nuclides in the mass region from 105 to 110 and greater than 150 will be greatly enhanced if Pu^{239} is present in the fuel sample. This is substantiated by the experimental data shown previously. Similar conclusions may be drawn from an examination of fission-yield ratios for fast fission vs thermal fission in both fissile nuclides. Thus from a determination of the relative abundances of selected fission product nuclides, information can be drawn about the Pu/U ratio and the energy spectrum of the reactor.

Additional information may be gained by considering the importance of the secondary production of nuclides by neutron capture. To illustrate this let us consider a few example cases. The simplest case is that of a nuclide like Cs^{137} . This isotope is produced by simple decay of the mass-137 chain to a long-lived isotope. In this case the capture cross section is quite small and the concentration of Cs^{137} atoms is a direct measure of the number of fissions which have occurred in the fuel. Another situation is illustrated by the case of Cs^{134} . This isotope is produced by decay of the mass-133 chain (iodine $\rightarrow$ xenon $\rightarrow$ Cs^{133}) where the end product is essentially stable.

Fig. 6. Pulse-height Spectrum of Gross Fission Products from a U^{235} Sample. Sample irradiated to 5% burnup at a flux of 10^{13} n/cm²/sec. Sample cooled 180 days.

Fig. 7. Pulse-height Spectrum of Gross Fission Products from a U^{235} Sample.
Sample irradiated to 50% burnup. Cooling time 250 days.

Fig. 8. Pulse-height Spectrum of Gross Fission Products from Pu²³⁹ Sample. Sample irradiated to 70% burnup at a flux of 2×10^{13} n/cm²/sec. Cooling time 180 days.

Fig. 9. The Ratio of Fission Yield for Various Mass Chains Plotted as a Function of Mass Number. Data are for ratio of $\text{Pu}^{239}/\text{U}^{235}$ (thermal fission).

The end product in this case (Cs^{133}) has a significant capture cross section which leads to the production of 2.3-yr Cs^{134} . In this case the secondary process introduces a second flux term in the mathematical expression for calculating the abundance of the final product. If we now consider the ratio of the concentration atoms of Cs^{137} to atoms of Cs^{134} , it should be directly proportional to reactor flux level. To demonstrate this relationship, we have plotted in Fig. 10 the results of a computer calculation of the ratio of Cs^{134} atoms to Cs^{137} atoms resulting from thermal fission as a function of reactor flux level. These calculations were made for 5% enriched fuel, and results are given for fission in both U^{235} and U^{238} . The calculations represent concentrations of these nuclides at the end of a 30-day irradiation. The slight downward curvature results from burnup of the fuel at higher flux levels. The measurement of the relative concentration of these two nuclides could then be used as an indicator of average flux for verification of fuel history in the reactor.

Fig. 10

Calculated Ratio of Cs^{134} to Cs^{137} Decay Rates in a Fuel Element as a Function of Reactor Flux Level. Data are for a 30-day irradiation period. Calculations include all pertinent reactions.

We may extend this type of process one step further. Let us consider the case of europium isotopes produced by capture following the decay of samarium isotopes. Of particular interest is the chain $\text{Sm}^{153} \xrightarrow{\beta^-} \text{Eu}^{153} \xrightarrow{n, \gamma} \text{Eu}^{154} \xrightarrow{n, \gamma} \text{Eu}^{155}$. In this case, the end product, 1.7-yr Eu^{155} , results from beta decay of the Sm^{153} to Eu^{153} which has a sizable capture cross section (320 b) which results in the production of 16-yr Eu^{154} . This nuclide in turn has a very large capture cross section (1400 b) for the production of 1.7-yr Eu^{155} . This three-step process introduces another flux term which results in the production of a nuclide whose concentration goes as the square of the flux when related to a first-order product such as Cs^{137} .

Figure 11 illustrates the possibility of using the ratio of the concentration of the nuclide Ru^{106} to Cs^{137} as a means of detecting the relative number of fissions from uranium and plutonium. These results indicate the ratio of these activities to be expected, for an irradiation time of 100 days, for fission in U^{235} and U^{238} plotted as a function of reactor flux. We see that the ratios are independent of flux and that the ratio differs by a factor of ten for the two fissile nuclides.

To summarize these possibilities, Fig. 12 shows the results of calculations for the concentration of a number of fission product nuclides. These results indicate the relative disintegration rates for Cs^{134} , Cs^{137} , Ru^{106} , Eu^{154} , and Eu^{155} as a function of irradiation time. The conditions selected for the computer calculations were 5% enriched fuel, a flux level of $10^{13} \text{ n/cm}^2/\text{sec}$, and a cooling time of 120 days. Examination of this figure

Fig. 11. Calculated Ratio of Activities in Reactor Fuel for $\text{Ru}^{106}/\text{Cs}^{137}$ as a Function of Neutron Flux. Calculation assumes irradiation time of 100 days.

indicates that the concentration of Cs^{137} is linear with irradiation time, indicating very little effect due to capture or decay. The behavior of Ru^{106}

Fig. 12. Relative Disintegration Rates of Five Selected Fission Products as a Function of Irradiation Time in a Nuclear Reactor. Calculations represent irradiation of 5% enriched fuel in a flux of 10^{13} . Cooling period of 120 days.

is similar, but long irradiation times result in some drop due to decay and/or capture processes. The Cs^{134} decay rate is seen to exhibit a much steeper slope because of the secondary capture process.

The slope of the Eu^{155} concentration is very steep, as one would expect from the higher-order processes.

Very briefly, we have attempted to convey some impression of the possibilities offered by the analysis of the gross fission product gamma-ray spectrum with high-resolution gamma-ray spectrometers. Possibilities include the determination of the fission ratio from uranium and plutonium, comparison of data from post-irradiation analysis with pre-irradiation data, measurement of residual fuel content, and to obtain information about the irradiation history of reactor fuels such as average flux level, reactor spectrum, and the measurement of the total number of fissions.

Although it is apparent that these techniques offer some definite possibilities, considerable experimental effort will be required to establish a realistic correlation between experimental results and calculations. In order to achieve a reasonable estimate of the capabilities of these techniques we have listed below some of the areas which need to be explored. We would propose that the most important limitation at the present time is the lack of high-quality basic information (e.g., fast-fission yields and precision values for capture cross sections of certain nuclides).

Proposed Areas for Study

A. Techniques

1. Extend performance of Compton-rejection annulus spectrometer to achieve improved response.
2. Establish performance of nondestructive techniques as related to well-established criteria.

3. Development of practical field instrumentation employing on-line computer data-processing techniques.

B. Basic Data

1. Extend accurate fission yield data to epithermal and fast regions.
2. Measure decay characteristics of selected nuclides.
3. Obtain improved primary and secondary capture-cross-section data for selected fission product nuclides and heavy elements.
4. Study gamma-ray branching ratios of selected fission product nuclides.
5. Study selected isotopic ratios as a function of nvt and reactor spectra.
6. Determine capture-to-fission ratio α as a function of reactor spectra.
7. Study of fuel being processed to obtain statistical verification of methods by comparison with measurements using nondestructive techniques.

THE NONDESTRUCTIVE ANALYSIS OF SPENT REACTOR FUEL BY GAMMA-RAY SPECTROSCOPY*

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I. INTRODUCTION

This paper reports the results and conclusions drawn from a series of studies carried out at MIT during 1964 and 1965. Much of this work was previously reported.¹⁻³ In this work a small Ge(Li) detector with an active volume of $\sim 0.6 \text{ cm}^3$ was used to detect the γ -ray spectra from spent fuel elements from the MIT reactor. The energy resolution of this detector was between 4 and 5 keV for energies of 0.5 to 2.5 MeV. Most of the measurements were carried out under water in the spent-fuel-storage pool, although the measurements by Brewer³ were done in air using the MITR fuel-transfer flask for shielding. The results from both measurements were essentially the same.

II. EXPERIMENTAL PROCEDURE

The measurements in the fuel-storage pit were carried out with a scanning device shown schematically in Fig. 1. The fuel was positioned 6 ft below the surface of the water, and the γ rays were extracted through an air-filled tube which had collimators at both ends. The collimator sizes used varied between $1/4$ and $1/16$ in. The fuel elements were the 16-plate MTR-type, fully enriched elements from the MIT reactor.

Fig. 1. Schematic Diagram of Apparatus for Gamma-ray Scanning of MITR Spent Fuel Elements.

A large number of fuel elements were scanned using the above device and their γ -ray spectra recorded. In Fig. 2 are shown the spectra from 4 elements of vastly different cooling times ranging from 1.5 weeks to $3\frac{1}{2}$ years. The principal fission product peaks are noted. The relative position of the curves to each other is arbitrary and was chosen only to show how the spectral features change with time. It is interesting to note that after a number of elements with different cooling times had been observed it became

*Much of the work reported in this paper was sponsored by the Air Force Cambridge Research Laboratories, Office of Aerospace Research under contract number AF19(604)-7492.

relatively easy for the spectrometer operator to look at the spectrum and guess the cooling time to within about 15%.

Fig. 2. Gamma-ray Spectra of MITR Spent Fuel as a Function of Cooling Time. Germanium-lithium drift detector No. 9-19 1; 3.5-mm depletion depth; 170-V bias; 77°K.

Curve	Fuel Element	Cooling Time
A	2M31	1-1/2 weeks
B	2M19	9 mo.
C	2M22	1 yr 6 mo.
D	2M1	1 yr 6 mo.

An enlarged view of the spectrum between 500 and 800 keV for an element with 9 months of cooling is shown in Fig. 3. The six fission products that have readily identifiable peaks in this region are indicated. It was observed that an element had to cool 4 to 6 weeks before the high background from short-lived fission products decayed enough to allow the peaks of these longer-lived products to become prominent.

The fission products Cs^{137} , Cs^{134} , and Zr^{95} were chosen for this analysis because the production of each had a distinctly different dependence on flux. In the case of the very long-lived (30 yr) Cs^{137} , the total amount produced is principally a function of ϕt , flux times time. On the other hand, Cs^{134} is produced by an (n, γ) reaction of Cs^{133} , the stable end product of the $A = 133$ fission product chain. Because its production is a two-step process

the amount present is roughly a function of $(\phi t)^2$. Finally, Zr^{95} is relatively short-lived (65 days) and so reaches saturation in the normal irradiation period of a year or so. The amount of Zr^{95} present after the irradiation is principally a function of flux ϕ alone.

The number of counts in peaks from these elements were determined with a computer code which subtracted a suitably averaged background from the total counts under the peak. The accuracy of the method was checked, and it was found that the overall peak areas were good to 5% or better in each case for the 9-month-cooled element. Figure 4 shows the results of a scan made along the length of a 9-month-cooled element. The increase at the bottom is due to flux peaking at the interface between fuel and moderator. No such peak occurs at the top, however, due to the presence of control rods which are partially inserted from the top during normal operation.

Fig. 3. Gamma-ray Spectrum of MITR Fuel Element 2M19 after 9 months Cooling. Germanium-lithium drift detector No. 9-19.2; 35-mm depletion depth; 1.6 cm²; 170-V bias; 77°K; run D4; 5/5/65.

Fig. 4. Relative Axial Distributions of Cs^{137} , Zr^{95} , (γ -724) and Cs^{134} (γ -605) Activities for MITR Fuel Element 2M19 after 9 months of Cooling.

III. METHOD OF ANALYSIS

In order to avoid the difficulties of an absolute calibration, it was decided to use ratios of the intensities of the fission products. The computer was programmed to calculate the ratio R_1 defined as the activity of Cs^{137} to the activity of Cs^{134} . Plots of this ratio as a function of neutron exposure for three different assumed fluxes are shown in Fig. 5. A similar plot for the ratio R_2 is shown in Fig. 6. These calculations require considerable nuclear data, including fission yields, half-lives, reaction

Fig. 5

Calculated Ratio of Cs^{137} Activity to Cs^{134} Activity in Spent MITR Fuel at Removal from Core as a Function of Neutron Exposure at Constant Flux.

Fig. 6

Calculated Ratio of Cs^{137} Activity to Zr^{95} Activity in Spent MITR Fuel at Removal from Core as a Function of Neutron Exposure at Constant Flux.

cross sections, and branching ratios. In addition, it is necessary to know the operating mode of the reactor, which in this case was on at steady power for 100 hr, then off for 68 hr, each week. The cooling time of the element is also required. It is not necessary to know the power level of the reactor, however. Corrections must also be made for detector efficiency and γ -ray self-absorption. However, these corrections are very small since all the γ rays have very nearly the same energy and it is only the ratio of these corrections which enters the calculations.

The solution of the equations is done graphically, as indicated in Fig. 7 which is a plot of flux ϕ versus exposure ϕt for the measured values of R_1 and R_2 . The intersection of the two lines gives the flux and exposure of that region of the element that was measured. A simple calculation also gives the actual time of irradiation.

Fig. 7. Curves of Neutron Flux vs. Neutron Exposure for Activity Ratios R_1 and R_2 Indicating Form of Solution for Flux and Exposure.

Table I gives the values of ϕ , ϕt , and t determined as described above for 14 different axial positions along element 2M19 which had cooled for 9 months. Note that as would be expected ϕ and ϕt vary with position along the element. The irradiation time, of course, should be identically the same for every position. The variations in irradiation time, therefore, provide an excellent check on the method. If any of the input data is in error, the values of τ will show a considerable variation. In this case, the mean value of τ agrees within $\pm 5\%$ of the value of τ determined from the operating log of the reactor.

TABLE I Summary of Results of Analysis of
Fuel Element 2M19 for Flux, Exposure, and Irradiation Time

Run	Position below Fuel Midplane, in	Neutron Flux ϕ , 10^{13} n/cm ² -sec	Neutron Exposure ω , n/kb	Irradiation Time τ , 10^8 sec
D4	-1	2.71	1.20	0.443
D5	0	2.61	1.17	0.446
D6	+2	2.62	1.21	0.461
D7	+4	2.61	1.26	0.484
D8	+6	2.42	1.08	0.448
D9	+8	2.31	1.19	0.513
D10	+10	2.23	1.17	0.525
D11	+11	1.72	0.84	0.488
D12	-2	2.61	1.20	0.461
D13	-4	2.50	1.09	0.436
D14	-6	2.30	1.03	0.448
D15	-8	2.29	0.984	0.430
D16	-10	1.93	0.848	0.439
D17	-11	1.54	0.738	0.479

Average Exposure $\omega = 1.08$ n/kb

Mean Irradiation Time $\tau = (0.464 \pm 0.008) \times 10^8$ sec

Actual Irradiation Time $(0.448 \pm 0.013) \times 10^8$ sec

From the measured exposure, the fission cross section, and the initial loading, it is possible to determine the burnup in an element. This

Fig. 8. Absolute Average Thermal-neutron Flux
Distribution within Fuel Element 2M19.

about the irradiation history of the highly enriched MTR plate-type fuel elements. It is possible to determine flux, irradiation time, and exposure, and then to calculate burnup to an accuracy of better than $\pm 10\%$. The method requires considerable nuclear data, which is one of the limitations to the accuracy.

was done and the agreement was well within 10% of the value determined by actual heat output. The flux values from Table I were compared to an independent measurement of absolute flux done by Mathews⁴ using cobalt wire, and the results are shown in Fig. 8. The agreement is especially striking since there is no normalization between the two sets of data.

IV. CONCLUSIONS

The results of these studies show that a gamma-scanning technique using a high-energy-resolution spectrometer can yield reliable information

In principle, the method could be applied to other types of fuel, e.g., low-enrichment oxide fuels from power reactors. However, a number of difficulties must be expected. These include the effect of fission product migration, the effect of plutonium fissions, self-absorption in these larger elements, and the much greater difficulty in handling and scanning large fuel bundles. None of these difficulties appears insurmountable as long as the cladding remains intact.

A number of improvements in the method are possible. These include taking advantage of recent detector developments, such as described by R. Heath in the previous paper, to obtain higher-resolution spectra. It should also be possible to extend the method to a considerably larger number of fission products and thereby obtain improved accuracy, as well as to get more consistency checks on the data. There is also the possibility that many investigators have speculated about of using fission products whose yield is different from plutonium and uranium fission to infer the ratio of uranium to plutonium fissions.

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DISCUSSION

DR. BEYSTER: I wondered about the use of the methods which were discussed in the last two papers. Suppose for some reason the power history is not known; the power history may be irregular and not steady, or has deliberately been varied. Will this be something that can ultimately be overcome so that you can find the quantities and isotopic composition of nuclear materials present in the fuel element, or must there be a pretty good idea of what that power history is?

DR. RASMUSSEN: The method we used assumes that you know the power distribution, which is an input to the code. If you give me the wrong power distribution, I am sure I could tell you it is the wrong one. We have done this on some elements that were removed from the reactor and replaced; when we put in the true history, we got a number that was reasonably good. But I don't think the equations can be solved backward for the way the reactor was operated, unless maybe you had many fission products to cross check.

DR. EDLUND: I have a question for Dr. Heath. Have you made any estimates as to how well your technique might work for measurement of the relative number of fissions of plutonium and U^{235} ?

DR. HEATH: I think we must color all answers by our lack of a lot of significant information. The information on fast-fission yields is in pretty sad shape. The dependence upon the distribution of particular fission product nuclides as a function of reactor spectra is a factor for which we suffer from a lack of basic information. However, given these data with sufficient precision, then I think we have a reasonable chance of doing what you would imply is possible. However, for this reason, really at the present time we cannot predict what the accuracy would be.

THE USE OF PHOTOINDUCED REACTION
PROCESSES FOR NONDESTRUCTIVE NUCLEAR
MATERIALS SAFEGUARDS APPLICATIONS

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The problems of nondestructive, quantitative isotopic identification of nuclear materials offer a real challenge to present-day technology. The forms in which nuclear materials may be found vary drastically in almost every imaginable way, including shape, size, and chemical composition. For these and many other reasons it is almost inconceivable that the problems of safeguards of nuclear materials will be resolved by any one unique method or process. It is furthermore evident to those that have wrestled with the problem that passive methods of identification are not entirely applicable enough really to resolve the many practical safeguards problems.

Active methods offer a method whereby an unknown assembly can be forced to react in its own characteristic manner to external forcing functions. One obvious advantage of active techniques is that the nuclear materials present are being examined directly, whereas in passive procedures an activity that has been produced over a long irradiation history is studied.

Our group at General Atomic has been studying one facet of the safeguards problem for a number of years. Clyde Jupiter will discuss the present program and the unique approach used tomorrow. This approach involves the measurement of the reactivity of the nuclear material and is especially useful for classification of appreciable quantities of material. Although our present study has not involved applications in the nuclear-material fuel cycle, the reactivity technique definitely has relevance here and should be evaluated insofar as the quantitative precision of measurement attainable in specific applications.

The purpose of my talk is to discuss the use of photoinduced reaction processes for nondestructive safeguards applications to nuclear materials. It is instructive at the beginning to mention a few key characteristics which are desirable in any safeguards interrogation system for nuclear materials (confining our attention to active methods of interrogation). These include:

1. the intensity of the induced effect;
2. distinctive signatures for the various nuclear materials;
3. overall simplicity, portability, safety, utility, and cost of the external source used for interrogation.

Although the above considerations are not all inclusive they are extremely important and will constitute the main subject matter for this discussion. Where possible, comparisons of the properties of the photoinduced reaction technique will be made with other proposed methods of active interrogation, such as those using pulsed fast-neutron irradiation.

Let us next consider the matter of relative intensities of photo-induced reactions attainable with the class of small electron accelerators envisaged for safeguards applications. The relative intensity of the effects induced in nuclear materials by an external source of radiation depends on at least four quantities (which are not totally unrelated): a) radiation source strength, b) solid angle subtended by the material at the source, c) penetration of the source radiation, and d) the probability for producing the identifiable signature in the nuclear material. A complete discussion of all these factors is beyond the scope of this paper. However, general order-of-magnitude studies easily illustrate the fact that with relatively simple radiation sources, signatures produced by photoinduced reactions can be higher by many orders of magnitude than those induced by other sources. For many of the signatures which we view as important the fission process is invoked and serves as a good basis for comparison.

For comparative purposes let us select three advanced typical fixed-energy (14 MeV) sources, both pulsed and steady-state. In Table I the possible modes of operation of these devices are intercompared in terms of fission yields obtainable at 3 in. from the source in one gram of uranium. The electron accelerator used is a present state-of-the-art commercial device. None of the 14-MeV sources given is made commercially as yet, but with a sufficient financial incentive might be developed. The Schmidt-Karlsruhe pulsed accelerator is the closest to being a really reliable long-running device free of typical 14-MeV target life and contamination

TABLE I Fissions Produced in 1 g of Uranium
at 3 in from Various Radiation Sources

Source	Total Fission Yield U^{238}	Pulsed Fission Yield U^{238}		
		1000 pps	200 pps	1/2 min
1. Pulsed Electron Accelerator (γ , fission)				
Internal Converter	7×10^9 fissions/sec	7×10^8	7×10^9	3×10^5
External Converter (20 MeV)	$\sim 10^{13}$ fissions/sec	10^{12}	10^{13}	5×10^8
2. Steady State (n, fission)				
14-MeV (d,t) Source (5×10^{11} n/sec)	2×10^6 fissions/sec	200	2000	~ 200 -2000
3. Pulsed 14-MeV Source (n, fission)				
Schmidt	1.6×10^6	-	1.6×10^6	80
Mather	4×10^4	-	-	4×10^4

problems. We have been very generous in the yields attributed to the 14-MeV neutron sources, and it can be seen that the fissions produced fall short of those which one can make with a present-day electron accelerator by many orders of magnitude.

In considering other, more typical practical applications, the four quantities (a through d) listed above may be commented on relative to this example. By far the most important factor in determining the overall high relative intensity produced by the electron accelerator is the accelerator source itself. To understand this one only needs to recall the very high bremsstrahlung intensities produced by 20-MeV electrons in heavy metal. This source is highly directional, in contrast to neutron sources, and thus can be concentrated directly on the nuclear material to be examined. This is especially true if the electron beam impinges on the uranium, as illustrated in Table I for "external converter." The nuclear material thus serves to produce its own bremsstrahlung beam, which subsequently photofissions or photoinduces reactions in the nuclear material.

Solid angle (quantity b above) is obviously also a factor in determining the much larger fission yields produced by electron accelerators. If the sources were located farther from the material to be examined, the advantage of the electron accelerator over the other sources in Table I would increase. It would almost always be possible to bombard the nuclear material with the electron beam directly, so that the external converter yield given in Table I need not change as the primary source is moved farther away, whereas the inverse-square law applies a rapid attenuation factor to most neutron sources.

The ease with which the high-current beam for an electron accelerator may be piped to experiments is well-known and is illustrated in Fig. 1, where some 12 radiation facilities distributed over 150 ft are shown.

Penetration of the source radiation (quantity c listed above) is often given as the primary factor in selecting a source for safeguards applications. As we have illustrated this is but one of many important factors and taken alone may be very misleading. What is meaningful, of course, is the number of reactions produced (in our example, fissions) at the desired place in our sample. In extrapolating from our example (Table I) to very large and thick samples containing nuclear materials we must concern ourselves with the mean free paths of bremsstrahlung X rays on the one hand and 14-MeV neutrons on the other to compare relative reaction rates. In unit-density (light) materials the mean free path for bremsstrahlung is about 20 cm, whereas for 14-MeV neutrons it is 10 cm. In heavy materials the situation is reversed such that the mean free path for 14-MeV neutrons is about 3 cm and for bremsstrahlung is

Fig. 1. General Atomic Linac Facility Showing the Large Number of Target Areas to Which Electron Beam Can Be Piped.

between 1 and 1.5 cm. One must thus be very careful in comparing intensities of signatures produced deep in bulk materials by 14-MeV neutrons with those photoproduced by bremsstrahlung. Over and above the unattenuated source intensity incident on the surface of the material one must factor in the relative attenuation mean free paths, which for many practical applications would appear to be about the same from the two sources differing by a factor of two or so either way, depending on the exact material to be penetrated. Another point worth remembering for interrogation deep in bulk material is that the primary source produces copious quantities of secondaries with a strongly degraded energy spectrum in the penetration process, and in many situations it will be literally impossible to pick up a signature that is associated only with the energy spectrum of the primary source. This is especially true when the nuclear material multiplies.

Regarding the primary cross sections for producing identifying signatures (quantity d above), again we are faced by the problem of becoming very specific. Generally, cross sections for photoinduced reactions and 14-MeV-induced reactions are of the same order; this factor, as in the case of the penetration factor mentioned above, is really not as important in determining the overall reaction rates attainable as the incident source strength (quantity a) and the material solid angle (quantity b). In summary, therefore, it would appear that intensity considerations as shown in Table I show that the reaction rates produceable in nuclear materials with electron accelerators are higher by several orders of magnitude (up to 10^7 in some cases considered) than those produced with typical high-energy neutron sources. Thus any active safeguards method of identification which depends on the production of fission in nuclear materials can be carried out with more overall source intensity by means of the photofission process. A very important point to observe is the advantage of direct electron impingement on the fuel to be examined. Intensity is, of course, not only important from the standpoint of producing meaningful fission rates deep in material in some cases, but in other cases may make it possible to interrogate fuel and obtain the requisite signatures faster and at more sampling positions.

A key characteristic to consider after intensity is the matter of obtaining very distinctive signatures for the various nuclear materials present in our unknown sample. Here again we will only endeavor to outline the very many possibilities which exist for producing meaningful signatures from photoinduced reactions. It is our belief that the characteristic signatures should differ between nuclear materials by as much as possible to increase the sensitivity and precision of the active interrogation process. No signature involving a difference of less than a factor of 2 would seem to be useful at all.

High-energy photons generally can interact with the material under study in a very wide variety of ways. Generally these interactions result in (1) photofission, (2) photoreactions other than fission: (γ, n) , (γ, p) , (γ, n, p) , $(\gamma, 2n)$, (γ, n, f) , (γ, γ') , and (3) photoactivation, namely, the excitation of characteristic residual induced activities.

Table II indicates the number of characteristic processes, considering only photofission, that are capable of being investigated to devise signatures of the fissionable isotopes. First, all materials of interest in safeguards applications are photofissionable, and the observables depend on the excitation energy provided by the external radiation source. Complete information is not available on the photofission process, since only a small amount of work has been conducted in the last 10 years. The time scale in the fission process is shown in Fig. 2. In contrast to neutron fission the threshold for photofission depends on photon energy for materials of interest, whereas the neutron fission of Pu^{239} and U^{235} is exoergic. Thresholds for photofission vary by 0.5 MeV and may be useful as signatures. The photofission proceeds near threshold in a very characteristic manner. An abrupt rise in cross section occurs at 0.5 MeV. Then the cross section rises much slower. The fission fragments separate at 90° to the electron beam in some cases, the prompt neutrons following the fragments, producing a very anisotropic and characteristic prompt-neutron angular distribution. Of course, the delayed neutrons do not follow the fragments. It may thus be possible to use the ratio of prompt to delayed neutrons near thresholds to identify the materials. The spectrum of prompt neutrons varies with angle because of this threshold center-of-mass effect. It is possible that the differences in photofission between nuclear materials can be emphasized at threshold since high excitation (gamma or neutron) tends to fill in the valleys in the fission-fragment mass-distribution curves and thus make the end products from fission of all materials more nearly the same. The absolute photofission cross sections for the various nuclear materials tend to differ by a factor

TABLE II. Distinguishing Characteristics of Photofission Process

1. Threshold energy
2. Threshold photoneutron energy spectrum
3. Threshold photoneutron angular distribution
4. Prompt-neutron yield vs photon energy
5. Prompt-neutron multiplicity vs photon energy
6. Prompt-gamma ray yield and spectra
7. Delayed-neutron yield and spectra vs photon energy
8. Delayed-gamma ray yield and spectra vs photon energy

Fig. 2. Graphic Representation of the Fission Process.

of ten at the giant resonance. Above threshold, the prompt-neutron yield and multiplicity changes drastically with photon energy. For U^{238} this variation is at least a factor of 100. An apparatus used for measuring this neutron yield variation is shown in Fig. 3.

Fig. 3. Experimental Layout for the Neutron-yield Measurements on General Atomic Linac.

The multiplicity of prompt neutrons (the number per fission) also changes with energy. Prompt-gamma ray yields from fission also vary with photon energy, and characteristic structure (not just X rays) has been observed in the spectrum at LRL. Data above 7 MeV may be very important. Delayed gamma rays from isomeric states produced in the fragments are one of our best indicators of characteristic fission processes. Discrete lines following 3 characteristic periods have been observed at times greater than $10 \mu\text{sec}$. The relative abundances of the delayed gamma rays differ by about a factor of 10 between U^{238} and U^{235} . These statements are illustrated in Figs. 4 and 5. The relative abundances of these isomers may also vary with the photon energy. In addition, delayed-neutron fractions and spectra have been shown to depend on photon energy and this behavior is also possible to incorporate into signatures for the materials involved. Yields per fission differ by a factor of 10 according to 1965 data of Nikotin.

Fig 4. Absolute Intensities of Gamma Rays as a Function of Time after Photofission of U^{235} , U^{238} , and Pu^{239} .

Fig. 5. Absolute Intensities of Gamma Rays from Photo- and Neutron-induced Fission of U^{235} and Pu^{239} .

We have only been able to indicate briefly the possibilities of developing signatures for nuclear materials based on the photofission process. It is clear that promising signatures are also possible through use of threshold photoreaction processes (other than fission) and the photo-excitation of characteristic longer-lived radioactivity. In the former case one might look at characteristic low-energy neutron groups produced by photo-threshold excitation by (γ, n) or other photo reactions in the nuclear materials. In the latter case one might measure short-lived fission product gamma rays, such as those of rhodium, which may differ drastically in abundance from plutonium to uranium fission. In conclusion, it is desirable to emphasize the vast number of differences that exist between photoinduced reactions in the various fissionable materials and the possibilities of turning these differences into meaningful signatures using variable-energy bremsstrahlung sources.

The third key characteristic which is desirable in an active interrogation system is an optimum radiation source. The source should be reasonably small, compact, portable, safe, relatively easy to operate, and inexpensive. This is not a simple task to accomplish and still preserve high intensities. Briefly let us review the electron accelerators available for active interrogation. There are at least 9 types of electron accelerators which vary in size and complexity. Traveling-wave Linacs, standing-wave Linacs, Betatrons, Van de Graaffs, ICT's, Dynamitrons, FFAG Betatrons, Microtrons, and Flash X rays are all candidates for this application. The devices preferable for safeguards purposes could be selected from this array and should definitely be of the variable-energy class. This energy variability is a key point and adds much flexibility to the methods and signatures available for interrogation. It is desirable to cover the range of electron energy from 4 to 20 MeV. All of these accelerators can deliver relatively high currents and all may be made small, compact,

portable, and easily serviced for field operations. Traveling-wave accelerators are in an advanced state of development. Figure 6 shows a typical small commercial linac. Our preliminary investigations indicate that the machines being considered are similar to accelerators used for cancer therapy, which are now much in vogue at our major medical centers and are free of complexity and operator problems, as well as of peculiar shielding and contamination problems. The optimum electron accelerator for material safeguards studies need not necessarily be more complicated than those presently contemplated for medical therapy or radiographic applications. This factor will, of course, reflect itself in a low or moderate cost of the accelerator installation and operation.

Although we will not demonstrate it here, it nevertheless seems promising that measurement of isotopic compositions of nuclear materials using photoinduced reactions is within the realm of possibility for clean fuel. This appears to be true simply because of very high source intensities and the very many signatures for the various fissile materials which can be devised from the prompt- and delayed-neutron and gamma emissions, and their dependences on photon energy. However, what is not so clear is that one can hope to observe useable intensities from photoinduced fission (for example) above the intense backgrounds from a spent fuel element. It is thus instructive to see how far one might hope to push the application of the electron-accelerator method. A technique one might attempt to exploit would be to irradiate a spent fuel element with a small accelerator electron beam directly and view the irradiated area with a small-aperture collimator. For example, if we viewed a spent Dresden fuel element about 100 days after shutdown we would find the total gamma power for the element to be of the order of a few kilowatts, but our gamma-detector-collimator arrangement would select only a 1/2-in. by 1/2-in. area at most of the element to view at one time. The area we view need not be larger than (but should be less than) the volume irradiated by the electron beam from the accelerator. The background source strength for this area would be 1.4×10^{13} MeV of fission product gamma rays per second. Using a 20-MeV electron beam one can produce in this volume 10^{11} fissions per electron-beam pulse. Suppose now one wants to use the relative intensities of various delayed γ -ray periods to construct a simple nuclear-materials signature. This would seem to be promising, for example, since the ratio of the 54- μ sec to the 32- μ sec periods for Pu^{239} is 1.15 and for U^{235} is 2.2. One then must view the fuel element for times like 100 μ sec with a high-resolution gamma-ray detector, looking at the time dependence of 850-keV or 250-keV lines for the 54- μ sec period, and 990-keV or 710-keV lines for the 32- μ sec period. During the 100- μ sec sensitive time of the detector the background intensity would be 1.4×10^9 γ -MeV per pulse (confined to a certain spectrum) while the signal intensities would be 10^9 isomeric gammas in the characteristic discrete-line spectra for the delayed gamma rays from fission. Thus it would appear then even in this extreme example that signal-to-background

Fig. 6. Simple Commercial Multipurpose Electron Accelerator (Varian).

ratios for active interrogation procedures based on electron impingement would be satisfactory. One, of course, can hope to develop other procedures based on photoinduced prompt- and delayed-neutron emission for spent fuel where the gamma background would not be so troublesome. In this regard photofission-threshold techniques appear to be extremely powerful, since the delayed-neutron fraction from the fissile nuclei all change markedly with photon energy near threshold and, of course, the prompt emission even has a very pronounced angular dependence near threshold. A lead-shielded spent fuel element could be studied with neutron detectors by measuring the ratio of prompt to delayed neutrons vs electron (photon) energy; each fissile material will have its own characteristic behavior with angle and photon energy. It would seem possible to resolve the composite behavior into the contributions from the major fissionable materials present since they behave so differently. Thus the isotopic ratios may be determined.

Of course, at this stage it is not possible to specify the best active method (based on photoinduced reactions) to use since so much of the basic photonuclear physics data are incomplete for nuclear materials. Much more basic experimental work needs to be done; feasibility studies of the various possible techniques and signatures need to be performed from the standpoint of possible precision attainable, and laboratory tests of possible safeguards procedures need to be made to verify feasibility. It would seem, however, that a system in which the total prompt- and delayed-neutron emission, and the time-dependent isomeric γ -emission spectra are studied at a series of incident photon energies from threshold would ultimately permit accuracy and even redundancy in absolute isotopic determinations. It is our hope to be able to embark on this proposed study of nondestructive interrogation of nuclear materials using active testing methods based on photoinduced reaction processes.

NONDESTRUCTIVE DETECTION, IDENTIFICATION, AND ANALYSIS OF FISSIONABLE MATERIALS

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Basic Technical Requirements in Nuclear-safeguards Implementation

The most important single technical requirement for nuclear-safeguards implementation is a nondestructive, direct physical means of detecting, identifying, and quantitatively analyzing fissionable materials in unknown mixtures of fissionable and nonfissionable materials. Four prime requisites of such a physical means, or technique, are:

- (1) high "penetrability," for complete assay capability throughout the entire volume of high-density bulk media (e.g., fissionable material in various bulk forms, such as fuel elements, ingots, slugs, slurries, melts, and process solutions);
- (2) environment insensitivity (to enable operation in the presence of extremely high radiation fields, extraneous or inert bulk materials, various perturbations, background "noise," spurious signals, etc.);
- (3) high "isotope discrimination power" (to distinguish unequivocally and quantitatively between the various fissile and fertile species in a given unknown bulk mixture;
- (4) the technique(s) should be nondestructive, accurate, and reliable; the equipment relatively simple, compact, and adaptable to various applications (e.g., direct, on-line isotopic assay in fuel reprocessing, fuel fabrication plants, and fuel handling and shipping facilities).

Of all the conceivable approaches to this basic problem in safeguards technology, it appears that techniques based on detailed analyses of delayed-neutron kinetic response to fast-neutron irradiations (pulsed, modulated, or DC) offer the greatest promise of meeting the very stringent requirements just outlined. All of the techniques of delayed-neutron kinetic response described herein utilize the characteristic differences in relative group yields and energy spectra of delayed neutrons from the various fission species to provide a sensitive, unique means of discrimination between the various fission species.

Methods of Delayed-neutron Kinetic Response for Nondestructive Detection, Identification, and Analysis of Fissionable Materials

The decay of delayed-neutron activity¹ following neutron-induced fission of various fission species (isotopes) has been measured and analyzed into six major groups based on their decay periods, which range from ~0.2 to ~55 sec. Although the six group periods are similar (but not identical) for all the major species undergoing neutron-induced fission, marked differences have been observed² in relative group abundances and energy spectra of delayed neutrons from the major fissioning species.

By detailed examination of the mass and charge distributions of the major fissioning species, these delayed-neutron-yield and spectral differences have been correlated² with the known variation of average fission chain length from one fissioning isotope to another. Thus the longer average β -decay chain lengths in fission of species with a high neutron-to-proton ratio (e.g., U^{238} and Th^{232}) account for increased yields of particular (especially short-period) delayed-neutron precursors which exhibit minor or negligible yields in fission of the species of shorter chain length ("low" neutron-to-proton ratio), such as U^{233} and Pu^{239} . This "chain-length effect" is most apparent among the short-period delayed-neutron groups, i.e., the increase of fission chain lengths in the sequence: U^{233} , Pu^{239} , Pu^{240} , U^{235} , Th^{232} , and U^{238} fission, correlates directly with increasing absolute yield of the fifth and sixth (shortest) delayed-neutron groups as one progresses through the sequence from U^{233} to U^{238} (cf. Tables 4-7 and 4-15 in Ref. 2).

These differences of delayed-neutron yield (as well as associated spectral differences to be discussed later) are uniquely characteristic of the individual fission species, and can be exploited to provide a unique method for nondestructive detection, identification, and analysis of individual fission isotopes in unknown mixtures of fissionable and non-fissionable materials.

In essence, the major technique described herein involves pulsing a system or sample of unknown composition with an intense, "instantaneous" pulse of neutrons (i.e., pulse duration short compared to all delayed-neutron periods τ_i) and analyzing, by special methods, the measured kinetic behavior of delayed-neutron activity following the pulse. Modulated or DC (i.e., saturation) irradiations may be similarly analyzed to provide complementary data to that obtained by the pulsed technique. As will be seen below, the short-pulse (short compared to all τ_i) irradiations generally provide optimum discrimination between the delayed-neutron response curves of the different fissioning species.

Analysis by Time Fiducial Methods

To outline briefly the analytical basis of the present method, we start with the delayed-neutron emission function $D(t)$,² which expresses the time dependence of delayed-neutron activity following an instantaneous neutron irradiation (irradiation time $T \ll \tau_i$) producing $\dot{F}T$ fissions in the irradiated sample:

$$D(t) = (\beta \bar{\nu}) \dot{F}T \sum_{i=1}^6 a_i \lambda_i e^{-\lambda_i t}. \quad (1)$$

Here $\beta \bar{\nu}$ (sometimes also designated n/F) is the absolute yield of delayed neutrons per fission, and a_i and λ_i are the measured abundances and decay constants of the delayed-neutron groups for a given fissioning species, respectively. We wish to compare the numbers of delayed neutrons emitted in various time intervals following both instantaneous and saturation irradiations. First we consider a single fission species; later the results will be generalized to mixtures of several different fission species.

The number of delayed neutrons emitted between $t = 0$ (end of irradiation) and an arbitrary time fiducial f following an instantaneous irradiation is

$$\int_0^f D(t) dt = (\beta \bar{\nu}) \dot{F}T \int_0^f \sum_i a_i \lambda_i e^{-\lambda_i t} dt = (\beta \bar{\nu}) \dot{F}T \sum_i a_i (1 - e^{-\lambda_i f}). \quad (2)$$

For the special cases of very short and very long* counting times, equation 2 reduces to

$$\int_0^f D(t) dt \rightarrow (\beta \bar{\nu}) \dot{F}T f \sum_i a_i \lambda_i \text{ for } f \ll \tau_i \rightarrow (\beta \bar{\nu}) \dot{F}T \text{ for } f \gg \tau_i. \quad (3)$$

It follows that the fraction R_f of delayed neutrons emitted in the interval $0 < t < f$ following an instantaneous irradiation is given by

$$R_f = \frac{\int_0^f D(t) dt}{\int_0^\infty D(t) dt} = \frac{(\beta \bar{\nu}) \dot{F}T \sum_i a_i (1 - e^{-\lambda_i f})}{(\beta \bar{\nu}) \dot{F}T} = \sum_i a_i (1 - e^{-\lambda_i f}). \quad (4)$$

*It may be noted that the case $\int_0^\infty D(t) dt \rightarrow (\beta \bar{\nu}) \dot{F}T$ forms the basis of the prompt-burst method of measuring absolute delayed-neutron yields.² This method, developed at Los Alamos, has been used to measure absolute delayed-neutron yields in fast and thermal fission of all the main fission species of practical interest.

For later use we introduce the complementary fraction

$$R_f^+ = 1 - R_f^- = \sum_i a_i e^{-\lambda_i f}, \quad (5)$$

which is simply the fraction of delayed neutrons emitted after time fiducial f (i.e., in the interval $f < t < \infty$).

Turning to the case of saturation irradiations, the emission function $D_S(t)$ (Ref. 2) represents delayed-neutron activity as a function of time following a saturation irradiation (irradiation time $T \gg \tau_i$) at constant fission rate $\dot{F}$:

$$D_S(t) = (\beta \bar{\nu}) \dot{F} \sum_i a_i e^{-\lambda_i t}. \quad (6)$$

Hence the number of delayed neutrons emitted in the time interval $0 < t < f$ following a saturation irradiation is

$$\int_0^f D_S(t) dt = (\beta \bar{\nu}) \dot{F} \int_0^f \sum_i a_i e^{-\lambda_i t} dt = (\beta \bar{\nu}) \dot{F} \sum_i (a_i / \lambda_i) (1 - e^{-\lambda_i f}), \quad (7)$$

which reduces to

$$\int_0^f D_S(t) dt \rightarrow (\beta \bar{\nu}) \dot{F} f \text{ for } f \ll \tau_i \rightarrow (\beta \bar{\nu}) \dot{F} \sum_i a_i / \lambda_i \text{ for } f \gg \tau_i. \quad (8)$$

The fraction S_f of delayed neutrons emitted in $0 < t < f$ following a saturation irradiation is given by

$$S_f \equiv \frac{\int_0^f D_S(t) dt}{\int_0^\infty D_S(t) dt} = \frac{(\beta \bar{\nu}) \dot{F} \sum_i (a_i / \lambda_i) (1 - e^{-\lambda_i f})}{(\beta \bar{\nu}) \dot{F} \sum_i a_i / \lambda_i}, \quad (9)$$

which reduces to

$$S_f \rightarrow \frac{(\beta \bar{\nu}) \dot{F} f}{(\beta \bar{\nu}) \dot{F} \sum_i a_i / \lambda_i} = \frac{f}{\sum_i a_i / \lambda_i} \text{ for } f \ll \tau_i \rightarrow \text{unity for } f \gg \tau_i. \quad (10)$$

We now extend the previous definitions of R_f - and S_f for a single fission isotope to the corresponding functions $\overline{R}_f$ - and $\overline{S}_f$ for a mixture of different fission species,* denoted by the index q . Thus for instantaneous irradiations, equation 4 may be generalized as follows:

$$\overline{R}_f = \frac{\sum_q (\beta \overline{\nu})^q \dot{F}^q \sum_i a_i^q (1 - e^{-\lambda_i^q f})}{\sum_q (\beta \overline{\nu})^q \dot{F}^q}, \quad (11)$$

which is simply a weighted sum of the known delayed-neutron kinetic functions $\sum_i a_i^q (1 - e^{-\lambda_i^q f})$ for each isotope q , the weighting factor being the fractional delayed-neutron yield (or production) from each isotope. For the special case of very short f , equation 11 reduces to

$$\overline{R}_f \rightarrow \frac{\sum_q (\beta \overline{\nu})^q \dot{F}^q \sum_i a_i^q \lambda_i^q}{\sum_q (\beta \overline{\nu})^q \dot{F}^q} \text{ for } f \ll \tau_i, \quad (12)$$

which again is simply a weighted sum of the known functions $\sum_i a_i \lambda_i$ for each isotope. From measured delayed-neutron characteristics of decay for all the major fission species, precise numerical values of $\sum_i a_i \lambda_i$ and their propagated errors (as well as the first and second moments of the delayed-neutron decay constants and mean lives) have been determined recently for all major fission species (cf. Table 10-1 of Ref. 2).

For saturation irradiations, equation 9 may be generalized as follows:

$$\overline{S}_f \rightarrow \frac{\sum_q (\beta \overline{\nu})^q \dot{F}^q \sum_i (a_i^q / \lambda_i^q) (1 - e^{-\lambda_i^q f})}{\sum_q (\beta \overline{\nu})^q \dot{F}^q \sum_i a_i^q / \lambda_i^q}. \quad (13)$$

*In general the number of major fissioning species in a given system or device will not exceed 3 (or in rare instances, 4).

For the special case of very short f , equation 13 reduces to

$$\overline{S}_f \rightarrow \frac{\sum_q (\beta \overline{\nu})^{q_F} q_f}{\sum_q (\beta \overline{\nu})^{q_F} q \sum_i a_i^q / \lambda_i^q} \text{ for } f \ll \tau_i, \quad (14)$$

and the reciprocal of $\overline{S}_f$ is again a delayed-neutron-yield weighted sum of the known kinetic functions $\sum_i a_i^q / \lambda_i^q$.

Since the expression for $\overline{S}_f$ given in equation 13 does not lend itself to isotope-analysis applications, an alternative formulation is the ratio $\overline{S}_f^- / \Delta$ of delayed neutrons emitted in the time interval $0 < t < f$ following a saturation irradiation to those emitted in a fixed short time interval $\Delta \ll \tau_i$ following the irradiation:

$$\overline{S}_f^- / \Delta = \frac{\sum_q (\beta \overline{\nu})^{q_F} q \sum_i (a_i^q / \lambda_i^q) (1 - e^{-\lambda_i^q f})}{\Delta \sum_q (\beta \overline{\nu})^{q_F} q}. \quad (15)$$

We also introduce the complementary ratio $\overline{S}_f^+ / \Delta$ for delayed neutrons emitted after time fiducial f (i.e., in the time interval $f < t < \infty$):

$$\overline{S}_f^+ / \Delta = \frac{\sum_q (\beta \overline{\nu})^{q_F} q \sum_i (a_i^q / \lambda_i^q) e^{-\lambda_i^q f}}{\Delta \sum_q (\beta \overline{\nu})^{q_F} q}. \quad (16)$$

For these expressions have the desired tractable form of a weighted sum of known delayed-neutron kinetics functions for each isotope q , the weighting function being, as before, the fractional delayed-neutron production from each fissioning species.

Letting $[W]$ symbolize the process of weighting with fractional delayed-neutron production from each fission species present in a given mixture, the preceding development can be summarized very simply as follows:

For Burst Irradiations:

$$\begin{aligned}\overline{R_f^-} &= [W] \sum_i a_i (1 - e^{-\lambda_i f}); \\ \overline{R_f^+} &= [W] \sum_i a_i e^{-\lambda_i f}.\end{aligned}\tag{17}$$

For Saturation Irradiations:

$$\begin{aligned}\overline{S_{f^-/\Delta}} &= \Delta^{-1} [W] \sum_i (a_i/\lambda_i) (1 - e^{-\lambda_i f}); \\ \overline{S_{f^+/\Delta}} &= \Delta^{-1} [W] \sum_i (a_i/\lambda_i) e^{-\lambda_i f}.\end{aligned}\tag{18}$$

Limiting values of these functions for very small and very large values of f are tabulated below.

$f \ll \tau_i$	Quantity	$f \gg \tau_i$
$f [W] \sum_i a_i \lambda_i$	$\overline{R_f^-}$	1
1	$\overline{R_f^+}$	0
f/Δ	$\overline{S_{f^-/\Delta}}$	$\Delta^{-1} [W] \sum_i a_i/\lambda_i$
$\Delta^{-1} [W] \sum_i a_i/\lambda_i$	$\overline{S_{f^+/\Delta}}$	0
$f \left\{ [W] \sum_i a_i/\lambda_i \right\}^{-1}$	$\overline{S_f}$	1

Precise numerical values of $\sum_i a_i \lambda_i$ and $\sum_i a_i/\lambda_i$ have been determined

recently, together with their least-squares propagated uncertainties, for all the major fission species.² These data are included in Table I, giving first and second moments of the delayed-neutron decay constants and mean lives.

TABLE I Moments of Delayed-neutron Mean Lives and Decay Constants (Computed, with least-squares propagated uncertainties, from data of Table 4-7, Ref 2)

	U ²³⁵	U ²³³	Pu ²³⁹
$\sum a_1/\lambda_1$ (sec)	12 753 $\pm$ 0 1834	17 876 $\pm$ 0 2866	14 6445 $\pm$ 0 1698
$\sum a_1/\lambda_1$ (sec ⁻¹)	0 4353 $\pm$ 0 0109	0 3004 $\pm$ 0 0194	0 3888 $\pm$ 0 0126
$\sum a_1/\lambda_1^2$ (sec ²)	466 05 $\pm$ 12 643	804 07 $\pm$ 22 302	532 94 $\pm$ 10 957
$\sum a_1\lambda_1^2$ (sec ⁻²)	0 682 $\pm$ 0 0464	0 376 $\pm$ 0 065	0 564 $\pm$ 0 042
	Pu ²⁴⁰	U ²³⁸	Th ²³²
$\sum a_1/\lambda_1$ (sec)	13 467 $\pm$ 0 2624	7 6803 $\pm$ 0 1025	10 059 $\pm$ 0 1657
$\sum a_1\lambda_1$ (sec ⁻¹)	0 4426 $\pm$ 0 0255	0 7847 $\pm$ 0 0179	0 5169 $\pm$ 0 0165
$\sum a_1/\lambda_1^2$ (sec ²)	460 68 $\pm$ 11 636	219 10 $\pm$ 4 313	370 62 $\pm$ 8 965
$\sum a_1\lambda_1^2$ (sec ⁻²)	0 753 $\pm$ 0 111	1 712 $\pm$ 0 080	0 766 $\pm$ 0 057

The $\overline{R}_f$ (burst-response) functions clearly emphasize the shorter-period delayed-neutron groups and the $\overline{S}_f$ (saturation-response) functions emphasize the longer-period groups. This capability for selective emphasis of both extremes of the delayed-neutron-period spectrum provides two complementary time-fiducial methods for identification and analysis of individual fission isotopes. Intermediate cases (i.e., between pulsed and saturation irradiations) are clearly obtainable through the use of modulated neutron sources.

From the foregoing development it follows that measured values of $\overline{R}_f$ (and also of $\overline{S}_f/\Delta$ when desired) for various f values (time fiducials defining various time intervals) can be used to determine fractional delayed-neutron yields from each major fission isotope in a given mixture of fissioning species. In such an analysis a linear superposition of known

kinetic functions (e.g., $\sum_i a_i(1 - e^{-\lambda_i f})$ in the case of $\overline{R}_f$ measurements)

with unknown coefficients (fractional delayed-neutron yields) is obtained in each time interval. The number of simultaneous equations in these coefficients (i.e., number and range of time intervals) can be judiciously chosen to suit the precision of the experimental data and to optimize the sensitivity and accuracy of the analysis in a given situation.

To indicate generally the capabilities and accuracy of the time-fiducial method, we examine the dependence of R_f and S_f/Δ functions

on f . Computer calculations* of R_{f-} and R_{f+} versus f are plotted in Fig. 1 for U^{235} , U^{238} , U^{233} , and Pu^{239} . Each R_{f-} curve approaches its limiting value $f \sum_i a_i \lambda_i$ for small f . Relative magnitudes of R_{f-} values (and

R_{f+} values, as well) for the different isotopes are clearly an index of the sensitivity or isotope-discrimination power of the $\overline{R}_f$ method. Ratios of R_{f-} and R_{f+} values--"isotope-discrimination ratios"--as a function of f are shown in Fig. 2, where appropriate limiting values are again indicated for small f . It is seen that optimum isotope discrimination in R_{f-} measurements is obtained in the range of small f values (<0.1 sec, say), while optimum discrimination in R_{f+} measurements is obtained at the larger f values (>10 sec, say).

Fig. 1
Delayed-neutron Fractions for Burst Irradiations

Fig. 2
Isotope Discrimination Ratios for Burst Irradiations

*Calculations are based on experimental delayed-neutron decay data;² Los Alamos prompt-burst decay data for all the major fission species are available in computer-listing form from G. R. Keepin, LASL.

The ratios R_{f-}/R_{f+} for each species can be used to provide still greater discrimination between individual species, as will be discussed below. It may be noted here that such ratios [e.g., $R_{f-}(0.1 \text{ sec})/R_{f+}(50 \text{ sec})$] constitute a measure of the average delayed-neutron decay "slopes" (early to late neutron emission) for the individual species.

Computer calculations of the kinetic functions $\Delta \cdot S_{f+}/\Delta$ and $\Delta \cdot S_{f-}/\Delta$ versus f (cf. Eqs. 18) are plotted in Fig. 3 for U^{235} , U^{238} , U^{233} , and Pu^{239} . Appropriate limiting values, $\sum_i a_i/\lambda_i$, are indicated at large and small f . Ratios of S_{f+}/Δ values are plotted in Fig. 4. Ratios S_{f-}/Δ

Fig. 3
Relative Delayed-neutron Fractions for Saturated Irradiations

Fig. 4
Isotope Discrimination Ratios for Saturation Irradiations

are not plotted as they are generally too small to be of practical interest, i.e., their maximum limiting value is $\sum_i a_i/\lambda_i$ at large f , which is just the limiting minimum value of S_f^+/Δ ratios (at small f).

In the respective time regions of their maximum values, R_f^- and R_f^+ ratios emphasize completely different extremes of the delayed-neutron-period spectrum, and hence constitute essentially independent measurements. Accordingly, we multiply appropriate optimum ratio values (from Figs. 2 and 4) together to obtain "effective discrimination factors" (cf. Table II) for the two isotopes concerned. These factors are seen to be simply ratios of delayed-neutron-decay "slopes" as defined above for the individual species.

TABLE II Effective Isotope Discrimination Factors Obtained from R_f^- , R_f^+ , and S_f^+/Δ

Isotopes	R_f^- Ratio ($f = 0.1$ sec)	R_f^+ Ratio ($f = 50$ sec)	Effective Discrimination Factor (R_f^- and R_f^+)	S_f^+/Δ Ratio ($f = 50$ sec)	Effective Discrimination Factor (R_f^- and S_f^+/Δ)
U^{238}, U^{235}	1.75	1.88	3.30	2.18	3.80
U^{238}, Pu^{239}	2.0	2.3	4.60	2.60	5.2
U^{238}, U^{233}	2.56	2.85	7.30	4.0	10.2
U^{235}, Pu^{239}	1.12	1.23	1.38	1.19 ^a	1.33
U^{235}, U^{233}	1.42	1.52	2.15	1.82	2.60
Pu^{239}, U^{233}	1.28	1.23	1.58	1.52	1.94

^aAt $f = 20$ sec

It is noteworthy that an effective discrimination factor as large as 10 is obtainable between the uranium isotopes U^{238} and U^{233} , a factor of 5 between U^{238} and Pu^{239} ,* and a factor of over 3 between U^{238} and U^{235} . The smallest discrimination factor (~ 1.4) is obtained between U^{235} and Pu^{239} . It is clear from Figs. 2 and 4 that still higher discrimination factors between all combinations of isotopes in Table II (except Pu^{239} and U^{235}) are possible using f values larger than 50 sec. In some cases, however, the available delayed-neutron intensity may limit the statistical accuracy of R_f^+ or S_f^+/Δ measurements for $f > 50$ sec (cf. later discussion of experimental technique and accuracy of measurements).

Before any analysis of relative amounts of fissionable material can be carried out, one must first identify the major fission isotopes present in a given system. Consider first the response to fast-neutron (e.g., 14 MeV) pulsed irradiations. From Fig. 1 we see that the presence

*Similarly excellent discrimination is obtainable between U^{233} and Th^{232} ; the $\overline{R_f}$ and $\overline{S_f/\Delta}$ methods should find extensive practical applications to assay and burnup determinations in U^{238} - Pu^{239} breeders and converters as well as to Th^{232} - U^{233} systems. Also, the large effective discrimination factor between U^{235} and U^{238} recommends the present methods for rapid, nondestructive determination of isotopic abundances in enriched uranium fuel.

of U^{238} as a major fissioning species will be indicated by measured $\overline{R_f^-}$ values in excess of the corresponding $R_f^-(U^{235})$ by more than a few percent. The same conclusion holds if measured $\overline{R_f^+}$ is less than $R_f^+(U^{235})$ by more than a few percent. In specific numerical terms this means, for example, that U^{238} is present if measured $\overline{R_f^-} \gtrsim 0.045$ at $f = 0.1$ sec, or if $\overline{R_f^+} \lesssim 0.06$ at $f = 50$ sec. Similarly for saturation irradiations in Fig. 3, if measured $\Delta \cdot \overline{S_f^+}/\Delta$ is less than ~ 12 for $f \lesssim 0.5$ sec, then U^{238} is present as a major fissioning species. This same approach can be applied to identification of U^{233} as a major fissioning species in an unknown system. Thus typical measurements establishing the presence of U^{233} would be $\overline{R_f^-} \lesssim 0.035$ at $f = 0.1$ sec, $\overline{R_f^+} \gtrsim 0.085$ at $f = 50$ sec, or $\Delta \cdot \overline{S_f^+}/\Delta \gtrsim 15$ for $f \lesssim 1$ sec.

Identification of individual species can be carried out still more effectively on the basis of R_f^-/R_f^+ ratios (delayed-neutron-decay "slopes") for the individual species. Numerical values of these slopes (for the time fiducials 0.1 and 50 sec) are given in Table III.

TABLE III. Average Delayed-neutron-decay "Slopes" Characteristic of Individual Fission Species

Fission Species	$\frac{R_f^-(0.1 \text{ sec})}{R_f^+(50 \text{ sec})}$	Fission Species	$\frac{R_f^-(0.1 \text{ sec})}{R_f^+(50 \text{ sec})}$
U^{238}	2.06	Pu^{239}	0.458
U^{235}	0.626	U^{233}	0.290

Delayed-neutron-decay slopes, being a combination of R_f^- and R_f^+ values, provide a more sensitive measurable characteristic of individual fission species than

either R_f^- or R_f^+ values alone. In specific numerical terms, if the observed slope, $R_f^-(0.1 \text{ sec})/R_f^+(50 \text{ sec})$, of an unknown mixture of fissioning materials exceeds 0.7, for example, the presence of U^{238} is clearly indicated (cf. Table III). Similarly, the presence of U^{233} is indicated if the measured delayed-neutron-decay slope falls below ~ 0.44 . In systems where U^{233} is not present (which in fact applies to nearly all practical cases) a measured decay slope less than ~ 0.60 would indicate the presence of Pu^{239} .

From a practical standpoint, the actual degree of discrimination ultimately obtainable between fission species as similar as U^{235} and Pu^{239} (which exhibit the smallest difference in delayed-neutron kinetic characteristics) must be investigated experimentally. Many approaches are possible, and warrant further careful and systematic laboratory investigation under various practical conditions. For example, in systems in which the other major fission isotopes are identified, and there remains only the discrimination between U^{235} versus Pu^{239} ,* one can make several

*In certain inspection and surveillance applications it is not always necessary to distinguish in detail between Pu^{239} and U^{235} components in a given system, it being sufficient to determine the combined U^{235} - Pu^{239} (and U^{233} , if present) "special" material contribution relative to the U^{238} (or Th^{232}) "safe" material ($k_\infty < 1$) contribution in the system. Such an analysis is, of course, readily obtained through appropriate R_f^- and S_f/Δ measurements. (For further details, see later discussion of Practical Applications.)

measurements at well-chosen f values (or take detailed multichannel time-analyzer data; cf. subsequent section on time-analyzer techniques) and perform least-squares fits to the data assuming, in turn, the presence of Pu^{239} and then U^{235} . The standard deviation of the overall least-squares fit to the data could then be used as a criterion for identification of U^{235} versus Pu^{239} .

In systems where identification by delayed-neutron response alone is difficult, one may employ the supplementary technique of high-resolution gamma spectrometry for identification of individual isotopes (this auxiliary technique is discussed in a later section).

Th^{232} has been largely ignored here for several reasons. Firstly, Th^{232} has a fission cross section for fast neutrons (e.g., fission spectrum to 14 MeV) smaller by nearly an order of magnitude than the other major fission species. Secondly, with the exception of the new large thorium reactors, Th^{232} is simply not used as a fissionable material in practical fission systems and devices. Its future use will likely be confined to large breeder and converter reactors, where its presence should certainly be well known! Nevertheless, where Th^{232} is a major fissioning species (as in future Th^{232} - U^{233} -cycle breeder reactors), it can be readily identified and analyzed by the delayed-neutron-response methods outlined herein. For example, from Table I one obtains good discrimination ratios between U^{233} and Th^{232} of 1.72 and 1.78 (for R_f^- and S_f^+/Δ ratios, respectively, at small f); of course, considerably larger discrimination ratios are available from R_f^+ and S_f^+/Δ measurements at large f (cf. Figs. 2 and 4).

Separate neutron irradiations with subthreshold fast neutrons and with superthreshold fast neutrons can clearly be used to provide further discrimination between the fissile and the threshold-fissioning species (notably U^{235} , Pu^{239} , and U^{233} versus U^{238}). (Fortunately for isotopic analysis applications, the delayed-neutron parameters a_i and λ_i are not sensitive to the energy of the neutron-inducing fission.)

In highly heterogeneous systems, there could arise perturbations due to differential self-shielding of delayed neutrons from the different fission species; implicit in the present development is the assumption that a flat-energy-response detector "sees" all delayed neutrons from the system equally well. Fortunately we are dealing with fast neutrons, so the assumption of system "transparency" is generally good, and high-efficiency, "flat-response" neutron detectors are readily available. Of course, problems are bound to arise in practical applications of delayed-neutron kinetic-response techniques to various types of complex, heterogeneous systems, and a systematic program of experimental investigation of the full potential of these techniques applied to such systems is now in progress at Los Alamos.

We now consider, by way of illustration, representative experimental conditions for typical pulsed irradiations of a mixture of fissionable materials. Modern compact, portable, pulsed neutron sources are capable of producing time-averaged yields well above 10^{11} neutrons/sec using the $D + T \rightarrow n + He^4$ reaction. The newly developed "neutron flash tube," for example, can produce $\sim 10^{10}$ D,T neutrons in a ~ 10 -ms interval at any desired rate of pulse repetition. More intense pulsed neutron sources are being developed specifically for the delayed-neutron response techniques described herein, and some two orders of magnitude increase in neutron pulse yields are presently obtainable. Assuming reasonable (e.g., 10%) geometrical coupling factors between the source and the system, and assuming nominal 1% neutron detection efficiency (actually in multi-detector 4π counting geometries, 90% or higher overall efficiency is readily attainable), adequate counting statistics can normally be obtained in several minutes. In a given experimental situation, of course, the number of pulsed irradiations [as well as the type of geometry of source(s), system and detector(s)] can be varied as necessary to achieve desired statistical accuracy in each f-defined time interval.

With a "flat"-energy-response neutron detector, $\overline{R_f}$ values can be readily measured to considerably better than one percent statistical accuracy in reasonable counting times (usually minutes, as already noted). The basic kinetic functions R_f^- , R_f^+ , and $\Delta \cdot S_f / \Delta$ are known to an accuracy of a few percent, as may be inferred from the uncertainties on the limiting forms $\sum_1 a_1 \lambda_1$ and $\sum_1 a_1 / \lambda_1$.^{*} Values of $(\beta \overline{\nu})^q$ are also known to a few percent accuracy.² Therefore the relative number of fissions of each major fissioning isotope present in an unknown mixture can be determined directly and quantitatively by kinetic-response methods. Appropriate ratios of fission cross sections can then be used to determine the relative amounts of each major fissioning isotope present if this further information is desired.

As we have seen, the experimental techniques for measuring delayed-neutron kinetic response are simple, direct, and do not require expensive equipment. The method does not require absolute detector calibration or fission monitor counting (although in some cases fission ratio determinations, either calculated or measured, may be desirable to determine relative amounts of each major fission species present).

Since β decay is essentially forbidden for half-lives shorter than ~ 0.1 sec, delayed neutrons are not expected, or observed, to exhibit half-lives shorter than 0.1 sec. Thus delayed-neutron kinetic-response methods

* In Table I the least-squares propagated uncertainties in $\sum_1 a_1 \lambda_1$ range from 2.3% for U^{238} to 6.5% for U^{233} , with an average of $\sim 3.6\%$ for all isotopes. The uncertainties in $\sum_1 a_1 / \lambda_1$ range from 1.2% for Pu^{239} to 1.7% for Th^{232} with an average of $\sim 1.4\%$ for all isotopes.

offer the great advantage that all measurements are made in a time domain which is completely free of time-dependent perturbations due to prompt-neutron higher modes, thermalization and diffusion effects (confined to the submillisecond time region), neutron time-of-flight smear, etc. Further important advantages are: (1) "environment insensitivity" (to the presence of high radiation fields, inert and/or extraneous materials, etc.) and (2) the high "penetrability" of fast neutrons through bulk media, clearly of major importance in detection and fission isotope analysis applications.

Analysis of Multichannel-time-analyzer Data

Equations 17 and 18 can be generalized to an arbitrary number of time intervals for analysis of multichannel-time-analyzer decay data to obtain fractional delayed-neutron production in a given system. The fraction $\overline{R}_n$ of delayed neutrons emitted in the n th time channel ($t_{n-1} < t < t_n$) following an instantaneous irradiation is (cf. Eq. 11)

$$\overline{R}_n = \frac{\sum_q (\beta \overline{\nu})^{q_F q_T} \sum_i a_i (e^{-\lambda_i^q t_{n-1}} - e^{-\lambda_i^q t_n})}{\sum_q (\beta \overline{\nu})^{q_F q_T}}. \quad (19)$$

Similarly Eq. 15 is generalized to the multichannel case:

$$\overline{S}_{n/\Delta} = \frac{\sum_q (\beta \overline{\nu})^{q_F q} \sum_i a_i^q / \lambda_i^q (e^{-\lambda_i^q t_{n-1}} - e^{-\lambda_i^q t_n})}{\Delta \sum_q (\beta \overline{\nu})^{q_F q}}. \quad (20)$$

Thus, in general for q isotopes and $n \geq q$ time intervals, a set of $\overline{R}_n$ (or $\overline{S}_{n/\Delta}$) measurements give n equations in q unknown coefficients (fractional delayed-neutron yields). The number of time intervals, n , can be adjusted to suit the precision of the experimental data simply by appropriate grouping of individual time-channel data from the multichannel analyzer. In the usual case $n > q$, standard computer least-squares techniques can be used to obtain best-fitted values of fractional delayed-neutron yields from measured $\overline{R}_n$ (and $\overline{S}_{n/\Delta}$) data and the known delayed-neutron kinetic functions evaluated at each time channel. This more detailed method of computer analysis of delayed-neutron response offers attractive advantages, including some further improvement in isotope discrimination power. However, for many routine analysis applications the more straightforward $\overline{R}_f$ method may be generally preferred for its simplicity and operational utility.

Delayed-neutron Group-energy Discrimination

Based on calculated end-point energies* for the delayed-neutron emission spectra from identified fission product precursors, increased discrimination between various fissioning species can be realized by the use of energy-biased neutron detectors (or differential neutron-energy spectrometers) in the delayed-neutron response measurements outlined above. As expected from fission product β -decay systematics, the shorter periods for delayed neutrons are associated with higher end-point β energies, and therefore exhibit correspondingly higher-energy delayed-neutron spectra.² As noted earlier, these shorter-period groups exhibit much greater abundance in the long-chain-length (high neutron-to-proton ratio) fissioning species such as U^{238} and Th^{232} . The "early" delayed neutrons (e.g., those emitted before $t = 1$ sec, say) from these long-chain-length species will exhibit a markedly harder energy spectrum than the corresponding early delayed neutrons emitted from the shorter-chain-length species (e.g., Pu^{239} and U^{235}). Also the exceptionally low energy² of the longest (55 sec) delayed-neutron group can be exploited to emphasize characteristic differences in the 55-sec-group relative abundances among the different fission isotopes. Thus, the use of a suitably calibrated energy-dependent detector can provide additional effective isotope discrimination (in nonmoderated or lightly moderated systems) by increasing discrimination factors between the fissioning isotopes. Detailed measurements of individual delayed-neutron-group spectra for different fission isotopes are now being undertaken at Los Alamos. These spectral measurements--and, in particular, the new energy spectra of the shorter delayed-neutron groups ($i = 4, 5$, and 6)--will permit further refinement and extension of this supplementary technique for identification and analysis based on delayed-neutron-group energy discrimination.

In its most general (and ultimately most incisive) form, the detailed neutron response method described herein would involve the determination of delayed-neutron time-energy response matrices for each individual fission species. The measured composite response of an unknown system would then be analyzed (e.g., by computer methods of matrix inversion) into its component response matrices weighted with fractional delayed-neutron yields from each individual fission species. The solutions for relative fission rates, relative amounts of various fissioning isotopes present, etc., are then obtained, as usual, from known $(\beta\bar{\nu})^q$ and fission cross-section data.

Techniques Supplementary to Kinetic-response Methods

Auxiliary or supplementary techniques for identifying various fissionable materials in unknown mixtures following pulsed neutron irradiation

*Calculated maximum energies, $E_{n_{\max}} = Q_{\beta} - B_n$ (cf. Table 4-11 of Ref. 2), range from 2.04 MeV for Br^{87} (55-sec-delayed-group precursor) to 5.2 MeV for I^{140} (0.5-sec-group precursor) and 9.1 MeV for Br^{93} (predicted 0.2-sec-group precursor).

include high-resolution time-dependent gamma spectrometry (e.g., using Li-drifted germanium detectors in appropriate coincidence-anticoincidence arrangements) to observe well-resolved gamma lines uniquely characteristic of individual fission species. The irradiation, observation, and analysis of time-dependent delayed gamma spectra could very conveniently be carried out concurrently with the measurement and analysis of delayed-neutron response characteristics. In such high-resolution fission product gamma-energy spectra, one may take advantage of the large yield variations (from one fission species to another) of particular fission product activities to aid in identification of individual isotopes. The greatest sensitivity of fission product yields to Z and A of the fissioning species will be found in or near the rapidly varying portions of the fission mass and charge distributions. Though certain characteristic gamma lines can be valuable in some cases for establishing the presence of individual fission species (i.e., for isotopic identification), in general gamma spectrometry cannot compete with the neutron techniques described herein for quantitative analysis of different fissioning species in bulk systems (cf. very high gamma background in irradiated material and large gamma absorption and self-shielding effects versus the relative "transparency" of most bulk systems to fast neutrons).

Thus high-resolution gamma spectrometry can provide a useful supplementary isotope-identification technique which, when desired, can easily be carried out concurrently with delayed neutron kinetic response methods for identification and analysis of fissionable isotopes.

Practical Applications

Practical methods of detecting, identifying, and analyzing fissionable material are of obvious importance in mining and processing of atomic energy raw materials as well as in many safety, control, regulatory, and surveillance functions of the atomic energy industry at both the national and international levels. The high sensitivity of the nondestructive kinetic-response methods described herein for discrimination between different fissionable isotopes is perhaps best exemplified by the practical case of U^{238} - Pu^{239} systems where an effective isotope discrimination factor of ~ 5 can be obtained between Pu^{239} and U^{238} . (Correspondingly large discrimination factors are obtainable between U^{233} and Th^{232} .) Thus the present techniques are admirably suited to the assay of Pu^{239} (or U^{233}) in power reactor fuel elements or breeder blanket material, or to rapid nondestructive evaluation of fission isotopic content in irradiated fuel, during fuel reprocessing, fabrication, or any other key point in the fission-fuel cycle. Similarly, the large effective discrimination factor between U^{235} and U^{238} enables direct determination of isotopic abundances in enriched uranium fuel elements, ingots, slugs, slurries, melts, solutions, etc.

It is important to note that the delayed-neutron response methods described herein constitute a physical sensor of the different fissioning

species, and therefore can be calibrated (for given source, detector configuration) to give a direct indication of the absolute amount of each fissionable species present in an unknown system. Thus, for example, reactor fuel elements of different, but known, composition can be used to calibrate delayed-neutron isotopic sensor equipment, which can then give directly the absolute amount of each fission species present in fuel elements of the same type but of unknown isotopic composition.

In addition to applications of kinetic-response methods to identification and analysis of individual fission isotopes, a further application of considerable practical significance is outlined below. From the data in Figs. 1-4 and Tables I, II, and III it is clear that maximum discrimination is obtained between two major categories of fission material: the so-called "special" (fissile) nuclear materials, U^{235} , $Pu^{239(240)}$ and U^{233} on the one hand, and the "safe" (fertile) nuclear materials (notably U^{238} and normal uranium, with $k_{\infty} < 1$) on the other. This practical distinction between "special" and "safe" nuclear material is obviously important in the field of nuclear inspection, surveillance, and control, which is primarily concerned with the detection and identification of "special" nuclear material--without necessarily distinguishing, at first, between U^{235} , $Pu^{239(240)}$ and/or U^{233} . (More complete isotope analysis can of course be carried out subsequently by more detailed kinetic-response methods, if desired.)

The special relevance of kinetic-response methods to this important practical problem is clear from the simple fact that measured delayed-neutron response reveals immediately and unambiguously the presence of "special" nuclear material as distinct from all other materials, i.e., the unique, "telltale" delayed-neutron response from U^{235} , $Pu^{239(240)}$, or U^{233} , or any combination of these, cannot be duplicated by any other combination of materials (fissionable or nonfissionable). Thus in general, with kinetic-response data readily at hand it is essentially impossible successfully to conceal significant amounts of "special" nuclear material. This is not to say that a would-be violator could not contrive concealment schemes based on "neutron-tight" containment, but any container which is "black" to 14-MeV neutrons (as could be ascertained immediately by a simple neutron-transmission measurement) would certainly arouse suspicion, whereupon further investigation and analysis could be carried out. The implications here for both national and future international nuclear inspection and surveillance methods are clear.

Nuclear Safeguards Research and Development Program at the Los Alamos Scientific Laboratory

In view of the growing necessity for more comprehensive and effective domestic and international control of nuclear materials, the field of nuclear safeguards is destined to assume major importance in the era of widespread proliferation of nuclear power (and its inevitable concomitant proliferation of fissile materials) which lies just ahead. Based on extensive

experience and leadership in technical fields closely related to safeguards technology, LASL is uniquely qualified to provide national leadership in this new technical field of nuclear safeguards and control.

The Laboratory is already a major contributor in the important area of nuclear energy safeguards, i.e., the detection of nuclear detonations via Vela Hotel, Vela Sierra, and long-range-detection programs. Now, to provide national leadership in the technology of nuclear materials safeguards, a new Pulsed Neutron Research Group has been formed at Los Alamos to carry out research and development of several promising new approaches to the detection, identification, and analysis of fissionable materials and fission devices.

The technical program of the LASL Pulsed Neutron Research Group (N-6) includes measurements of the detailed kinetics and energy characteristics of delayed fission neutrons (as well as fission-product gamma emitters). Both basic research into delayed-neutron properties and the development of practical systems utilizing delayed-neutron kinetic-response techniques for detection, identification, and analysis (DIA) of fissionable material are necessary phases of this technical program. Some of the required basic measurements to be carried out by Group N-6 are:

- a. Absolute delayed-neutron yield at high incident neutron energy. (Existing yield and abundance discrepancies at high energies, up to 14 MeV, will be fully and unequivocally resolved.)
- b. Delayed-neutron-group abundances and decay constants for 14-MeV-neutron-induced fission of all major fission species (these measurements are required for optimum exploitation of delayed-neutron response methods).
- c. Energy spectra of individual delayed-neutron groups from the major fissioning species. Associated program of development of neutron spectrometer. (Investigation of increased isotope discrimination factors using energy-sensitive neutron detectors.)
- d. Extensive studies of the neutron-rich fission products, particularly delayed-neutron (and later delayed-gamma) emitters, with a view toward advanced DIA applications.

As nearly coincident as possible with the required basic measurements listed above, Group N-6 will apply delayed-neutron kinetic-response techniques to the development of practical DIA systems. This phase of the technical program will include studies of a variety of differential shielding (transmission) effects in representative types of systems, the development and/or optimization of high-efficiency "flat-response" detectors and differential neutron spectrometers to achieve maximum isotope discrimination. Empirical approaches to DIA will also include measuring

and cataloging delayed-neutron response "signatures" for various types of systems and devices, determining the ultimate sensitivity of DIA systems in identifying "special" (fissile) versus "safe" (fertile) fissionable materials, and identifying and analyzing unknown or clandestine systems. Advanced experimental and analysis techniques will be pursued through multidimensional, multichannel time and energy analysis, development of high-intensity pulsed sources, and the extension of delayed-neutron response methods to the determination of absolute amounts of fissionable materials present in unknown systems (using appropriate calibration techniques). Ultimately, hardware concepts will be implemented to develop an integrated source-detector-analyzer system with demonstrated practical performance for various types of inspection and surveillance applications.

The principal research tool of Group N-6 at present is a 350-kV pulsed Cockcroft-Walton accelerator which was recently installed in its present location at TA-18, Los Alamos. This versatile pulsed neutron source is operated in both the direct current and pulsed modes with a total neutron source strength in excess of 10^{11} neutrons/sec using the $D(T,n)He^3$ reaction. In the pulsed mode, the accelerator can produce neutron pulses of any desired width down to the nanosecond region (required for prompt-neutron measurements of fast metal systems) with corresponding pulse-repetition rates variable up to the megacycle range. A 5-kV beam-deflection system was recently developed for post-acceleration pulsing to supplement the existing pre-acceleration pulsing capability in the high-voltage head.

As part of the program of basic data required for the development of practical DIA systems, the absolute yields of delayed neutrons from high-energy (14 MeV) fission of the major fission species are currently being measured through use of the CW accelerator as the neutron source. The general experimental arrangement is shown in Fig. 5. The beam-acceleration tube can be seen extending from the high-voltage deck (containing ion source, pre-acceleration pulsing equipment, etc.) at the upper right to the vacuum cabinet, beam-deflection magnet, and thence through the 25° left beam port to the target and multiple detector configuration at the lower left.

The existing N-6 Cockcroft-Walton accelerator provides an adequate pulsed or DC neutron source for some of the basic measurements required in detection, identification, and analysis techniques, and for nuclear safeguards research in general. However, for the development of practical, high-efficiency DIA systems, a more compact, more intense source of neutrons is required. A comprehensive survey of existing and foreseeable sources has indicated that the Dense Plasma Focus (DPF) pulsed neutron source recently developed at Los Alamos³ is particularly well suited for DIA applications.

Fig. 5. Experimental Arrangement for Absolute Measurements of Delayed-neutron Yields

In the DPF device, stored magnetic energy is rapidly converted to plasma energy and then compressed by its self-magnetic field. Plasma pressures (nkT) of the order of 3×10^5 atmos are developed in a region of ~ 0.5 -mm diameter and 3.5 mm in length. The resulting neutron yield is $\sim 3 \times 10^{12}$ D,T neutrons in $\sim 0.15 \mu\text{sec}$, and is not inconsistent with the assumption of thermonuclear origin.

A 40-kilojoule DPF pulsed neutron source is presently being constructed by Group N-6 for use explicitly in its DIA program.

Other significant Los Alamos activities in support of the Laboratory's nuclear safeguards research program include:

- a. Computer studies aimed at mathematical simulation of delayed-neutron kinetic-analysis techniques in various types of composite systems (to provide guidance for experimental programs, establish precision requirements on measurements, etc.).
- b. DPF source design and development assistance from plasma physics and CTR people at Los Alamos.
- c. Radiochemical analyses for fission monitoring, yield normalization, etc.
- d. Development and preparation of radioactive sources with specialized characteristics.

- e. Mass-spectrometric and chemical analyses.
- f. Neutron-source-calibration services.
- g. Source radiography and activation services.

A portion of the safeguards research effort at Los Alamos is also concerned with the exploration of new ideas and different approaches to present and future technical problems in the field of nuclear safeguards.

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