Energy and Technology Review

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**AUTO-
MATED
URANIUM
ASSAYS**

MASTER

**LAWRENCE
LIVERMORE
LABORATORY**

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ABOUT THE COVER

A sample of uranium-bearing solution undergoing assay in a computer-automated analyzer developed at LLNL as part of the nuclear safeguards and security program. For better visibility, the protective plastic collar around the beaker has been removed. Precise control of the currents passing through the gold electrodes and accurate measurements of the potentials in the solution (coulometric titration) enable the analyzer to assay uranium with precision, speed, economy, and freedom from clerical errors. The edge of the beaker for the next sample to be assayed can be seen at the lower left. For a description of these automated uranium assays, see the article beginning on p. 5.

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Briefs

The short items on this page announce recent developments of importance. Some of these items may be reprinted in future issues; none of this material is reported elsewhere in this issue.

RENIUM-187: A NEW ASTROPHYSICAL CLOCK

The results from recent LLNL experimental measurements of two exotic nuclear reactions, combined with previous half-life measurements and well-understood stellar processes, place the age of the universe at 20 billion years. This is six billion years older than the age determined by the more familiar but less accurate uranium-thorium decay clock. The new age estimate is based on a fundamentally simple nuclear mechanism: the decay of radioactive rhenium-187 into its stable daughter osmium-187.

Measuring the age of the universe is not prompted by mere idle curiosity. Its age is a fundamental datum in most theories of cosmology—understanding how the universe came into being and what its eventual fate may be.

One attractive feature of the rhenium-osmium clock is that all the competing reactions that influence it can, at least in principle, be measured. By contrast, the production rates of uranium and thorium in stellar processes, i.e., supernovae, are unmeasurable and can only be estimated from theoretical considerations. Another advantage of the rhenium-osmium clock is its

ten-times-longer time scale. The half life of rhenium-187 is 44 billion years, more than twice the estimated age of the universe. This increases the accuracy of the age estimate by minimizing the influence of uncertainties in the details of the stellar processes involved.

When the rhenium-osmium clock was first proposed in 1964, it was recognized as being theoretically more accurate than the uranium-thorium clock. It could not be used at that time, however, because of two hard-to-measure neutron capture reactions that could affect the amount of osmium-187 and influence the estimated age. These are the reactions that have just now been measured.

The interfering reactions in question are the creation of osmium-187 from osmium-186 and the destruction of osmium-187 (by transmutation into osmium-185), both by neutron capture. Osmium-186 and -187 are rare isotopes; it was hard to get samples pure enough and large enough for the cross-section measurements. An Oak Ridge Calutron running for months provided a few grams of each isotope. The LLNL 100-MeV electron linear accelerator provided the intense source of neutrons that was essential for the cross-section measurements.

FORECASTING THE WORLD'S CRUDE OIL SUPPLY

A recent LLL study indicates the distinct possibility of a temporary oil price break in the early 1980's. The long-range scenario remains unchanged: diminishing crude oil resources and steeply rising prices followed by elimination of oil as a feasible energy source for high-volume markets (e.g., transportation and electrical production). Whether or not the world price of crude oil softens, we consider the continued development of alternative energy sources to be imperative.

The Federal Energy Administration's Project Independence report of November 1974 and its updated version, *The National Energy Outlook* of March 1976, outline ways in which independence from foreign energy sources may be possible by 1985. Many competent observers are skeptical; a common view is that herculean steps by U.S. agencies and government groups will be needed to achieve independence. Even with accelerated leasing schedules for oil and gas exploration, strong energy-conserving measures, relaxation of environmental standards, opening up of the several Naval Petroleum Reserves, and use of enhanced recovery techniques in oil fields, it appears almost certain that the U.S. will be importing oil and gas for at least ten years. Under these circumstances, availability of supplies, security of foreign sources, and prices of imports are all vital issues that intelligent planning in the energy field must take into account.*

Most projections assume that today's high crude-oil price is permanent. It is argued that economic recovery will restore the high rate of oil demand, that declining U.S. production and the loss of Canadian gas and oil will make us more dependent on Middle Eastern oil, and that any efforts to stockpile oil will further inflate demand. It is possible, however, to postulate a set of circumstances that will reduce oil prices in the early 1980's despite Western dependence on oil from the Organization of Petroleum Exporting Countries (OPEC). Although temporary, such a reduction could erode public support for energy research to develop alternative fuels. It is a prospect, therefore, that should be considered in national energy planning.

The circumstances leading to a drop in the crude

oil price have several components. These include a rapid expansion in worldwide crude oil production, a much-reduced annual growth in demand, and a pressure of rising expectations in the OPEC countries as they develop appetites for foreign imports and technology. Together, these circumstances could force oil exporters to compete for markets. Although outside the scope of this article, the political climate and social structure of the major OPEC countries are also pertinent.

Worldwide Oil Production

Continued and rapid development of worldwide crude oil supplies, including the vast resources of the U.S.S.R. and China, is a prime factor in determining the future price of oil. For one thing, oil found outside the OPEC is potentially competitive and may directly affect the artificial OPEC price. Even more important, however, it can minimize the new producing country's current dependence on imported oil. Partial self-sufficiency by importers in Latin America, Africa, and the Far East could reduce OPEC markets.

It is too early to know if this reduction will occur. All nations are emphasizing the exploration, development, and production of indigenous supplies. In some places—e.g., Alaska, Peru, Colombia, and Ecuador—pipeline construction currently presents bottlenecks. Within a few years these pipelines will be in service and supplying additional domestic oil. In Brazil, and offshore both Argentina and India, where exploration is only now under way, it may be some years before oil now listed as "undiscovered" can be tabulated among the reserves.

Figure 1 gives projections of world oil production over the next 75 years. The numbers in parentheses represent the total resource in each region, in billions of tonnes. The inset is a breakdown of the "Other" category, consisting of estimates of recoverable oil from Latin America, Africa, the Far East, Western Europe, Canada, and Antarctica.

The individual curves in Fig. 1 combine historical production data to 1975 with "surprise-free" projections of future production. Beyond 1985 the projections become more speculative, making the shapes of the curves somewhat arbitrary. They are constrained only by the known initial slope and by the area

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under each curve, which corresponds to the estimated total resource. The flat tops of the curves reflect the opinion that it takes a projected useful life of 20 to 25 years to justify the construction of new refineries and production equipment.

Figure 1 emphasizes one fact: the world's total oil resource is finite. There is only so much crude oil, and it is not being replaced. Conservation and enhanced recovery methods may stretch the supply, but they only postpone the final outcome. Long before it is all gone, oil will become far too expensive to burn. Alternate energy systems must be ready to fill the void.

Table 1 shows the world's reserves and resources of crude oil in somewhat greater detail. Proved reserves represent what is known with reasonable certainty to be recoverable from known reservoirs under existing economic and operating conditions. Prospective reserves represent an educated guess at how much more may be recoverable from these same reservoirs. Past experience shows that the proved-reserves estimate is generally conservative; five or six times more oil is usually pumped out of any new discovery than was originally thought to be there. It is common practice to combine the proved and prospective reserves into the single category of discovered oil.

Fig. 1. World crude oil production, based on actual production data and on projections described in the text. The numbers in parentheses represent total reserves and resources for each country. The inset shows components making up the curve labeled "Other" in the main illustration; note the expansion of the vertical scale.

Table 1. World crude oil reserves and resources

Region	Discovered (proved and prospective reserves), 10 ⁹ tonnes (10 ⁹ bbl) ^a	Undiscovered (potential resources), 10 ⁹ tonnes (10 ⁹ bbl)	Total, 10 ⁹ tonnes (10 ⁹ bbl)
Middle East	60.0 (435)	21.0 (150)	81.0 (585) ^b
Western Europe	3.3 (24)	7.8 (57)	11.1 (81) ^b
Africa	9.6 (70)	9.7 (71)	19.3 (141) ^b
Central and South America	5.3 (39)	11.2 (82)	16.5 (121) ^b
Far East	4.1 (30)	12.3 (90)	16.4 (120) ^b
Antarctica	-	2.7 (20)	2.7 (20) ^b
Canada	1.2 (9)	9.6 (70)	10.8 (79) ^b
United States	14.4 (105)	20.0 (145)	34.4 (250) ^c
U.S.S.R.	11.4 (83)	} 35.0 (256)	46.8 (342) ^d
Eastern Europe	0.4 (3)		
China	5.9-7.7 (43-56)	6.0 (44)	13.0 (94) ^e
Total	116 (848)	135 (985)	252 (1833)

^aFigures originally expressed in barrels (bbl) have been converted to tonnes with the average conversion factor of 7.3 bbl/tonne.

^bRef. 1.

^cRef. 2.

^dRefs. 1 and 3. The combined U.S.S.R. and China estimate¹ of 41×10^9 tonnes (300×10^9 bbl) has been reduced by 6.0×10^9 tonnes (44×10^9 bbl) to break out the China figure.

^eRefs. 4 and 5.

Resources, on the other hand, refer to oil that has not yet been discovered. Estimates in this elusive category are necessarily based on very subjective judgments; they differ widely for individual countries and for the world as a whole. Some estimate of undiscovered oil is important, however, because in many cases it is much larger than the proved and prospective reserves. Furthermore, it tends to determine the upper boundary of our limited oil supply.

Free-World Demand for Crude Oil

Historically, every increase in oil-production capacity has eventually been equaled by demand. Now, however, with the price artificially inflated, some customers are being squeezed out of the market. Many Third World countries lack the resources and foreign exchange to buy the oil they need for fueling their fledgling industries. Ambitious development plans have been set back or in some cases cancelled.

Even in this country the high price has had its effect. Last year's recession, following the oil embargo, produced a definite dip in our oil consumption. One index of the current economic recovery is our rising demand for petroleum products.

Forecasting future demand is difficult because

austerity measures and conservation practices enforced by government regulations can affect it. However, some estimates can be made. Even if oil prices remain constant, the growth of free-world population alone could force at least a 1.6% annual increase. Our estimates, based on continued high prices, place the average yearly increase in demand over the 1974 to 1995 period at 2.8%. (The increase during recovery from the recession may be double or triple this rate, but the increase around 1995 would be nearly zero due to the leveling off of conventional production.)

Continued high oil prices and a concerted effort to control consumption could produce a demand curve such as that shown in Fig. 2. This is based on a 5.5% annual increase during the recovery period of 1975 to 1980, a 1.6% increase from 1980 to 1985, a 1.7% increase from 1985 to 2000, and a 1.9% increase from 2000 to 2010. Such limited demand growth would be below the anticipated growth in world production capacity and would force additional cutbacks beyond the OPEC's present restrictions on output.

Rising Expectations

In addition to expanded world production and reduced demand growth, the third major factor controlling the price of oil is the economies of the

Fig. 2. Historical and projected Free World demand for crude oil assuming continued high prices. Economic or regulatory restraints are assumed to restrict the annual growth in demand to 1.6% in 1980-1985, 1.7% in 1985-2000, and 1.9% in 2000-2010.

OPEC producers themselves. Oil is basically their only source of income. Individually, many OPEC countries can raise all the money they need by pumping more oil; collectively, they can maintain a high price by restricting production. They can't all do both, however.

Before the oil embargo, when the price of oil was relatively low, most OPEC countries had modest budgets. Many have since embarked on ambitious development programs aimed at modernizing and industrializing their economies. Much of this commitment is based on future production. The question is not whether they will be able to pay for these programs, but whether they will be able to pay without increasing production. Substantial production increases may well mean lower prices.

There are signs that something of this sort is already happening. Iran, for example, is running a budget deficit. In the last few years, it has contracted for a \$500 million expansion of telephone facilities, a \$250 million wood-products complex, a \$550 million expansion of the Teheran airport, a total of \$1.5 billion in rail contracts, and another \$2 billion for nuclear power plants. By 1980 military equipment expenditures may reach \$5 billion per year and agricultural imports are expected to reach some \$2 billion annually.

Before the recession, OPEC members operated their oil facilities at about 82% capacity. During the recession and the mild winter of 1975, these same facilities apparently operated at little more than 70% capacity. In the 1980's, further production cutbacks could jeopardize some OPEC nations' internal security and domestic programs. There are indications that this 30% cutback, or "shut-in capacity," is about the limit, and that any further softening of demand would force one or more of the OPEC countries into price concessions.

Conclusions

The gist of the preceding analysis is that the current high price of crude oil is not necessarily permanent. Assuming that worldwide oil production outside the OPEC nations continues to expand, that economic or regulatory restraints can hold down the growth of demand, and that rising expectations within the OPEC nations will force them to continue their present spending trends, some adjustment in prices is a reasonable consequence. Extrapolating current trends, it appears that the price break could come about 1981. This outcome cannot be predicted with confidence, however, without a more detailed analysis of the three assumptions on which it is based.

A drop in crude oil prices, welcome as it might be from the consumer's standpoint, could have a serious impact on the nation's pilot studies to develop alternative energy sources. Most of these studies were originally justified on the assumption of continued high oil prices; a price drop might appear to contradict that assumption and thereby weaken public support.

Curtailling these projects would be short-sighted, however. The world's total oil resource is limited, and world demand will ultimately exceed the available supply. Any drop in crude oil prices will be temporary. Alternative fuels and energy sources must be actively pursued so that they may be ready to fill the clearly discernible demand gap that will open when oil production levels off.

Key Words: energy - forecasting; fossil fuels; Independence Project; oil - demand; petroleum resources; power resources - economics.

AUTOMATED URANIUM ASSAYS

Precise, timely inventories of enriched uranium stocks are vital to help prevent the loss, theft, or diversion of this material for illicit use. A wet-chemistry analyzer has been developed at LLL to assist in these inventories by performing automated analyses of uranium samples from different stages in the nuclear fuel cycle. These assays offer improved accuracy, reduced costs, significant savings in manpower, and lower radiation exposure for personnel compared with present techniques.

The analytical chemistry of nuclear fuels is becoming increasingly important as the commercial power reactor industry expands. At every stage of the nuclear fuel cycle from mining uranium to reprocessing spent fuel and storing the wastes there must be accurate assays of the quantities of uranium on hand. Especially in purified and enriched form,

uranium has both high commercial and strategic value. Precise, timely inventories are thus needed to safeguard against its loss, theft, or diversion for illicit use.

This adds up to a lot of assays. The manpower requirements alone are formidable. When one includes the possibilities of operator bias and simple clerical error that plague any manual procedure, and the amount of cross checking required to counteract them, he gains some idea of the magnitude of the task.

We have developed an automated, computer-controlled analyzer⁶ to assay uranium in solid or liquid samples taken from various stages in the nuclear fuel cycle. This system offers a significant saving in manpower, freedom from operator bias, and fewer clerical errors. Analyses are inexpensive and fast enough to permit replicate measurements, thus lowering the statistical uncertainty of the results. Extensive built-in checks assure the quality of the analytical results. As an added bonus, automatic operation exposes the operator to less radiation.

Contact Jack E. Harar (Ext. 7852) for further information on this article.

Fig. 3. Basic procedure for the assay of uranium materials with our computer-automated analyzer. The steps with broken outlines are semiautomatic; those with solid outlines are fully automatic. Weighings on an electronic balance replace volumetric measurements because of their greater accuracy and the ease of transmitting results to the computer memory. Pre-titration adjustments of the oxidation state, the titration itself, and the final calculations are all performed by the computer without human intervention.

Assay Procedure

The basic procedure for our automated uranium assay is outlined in Fig. 3. If the sample is solid, it is weighed and dissolved in an appropriate acid mixture. The resulting solution (or the original solution, if the sample is liquid) is weighed, and then a weighed portion, or aliquot, is taken for analysis. The aliquot is partially evaporated with sulfuric acid and placed on the automatic sample changer. The analyzer then automatically processes the aliquot through the rest of the procedure.

A computer controls the addition of four reagents, in metered quantities and in the right sequence, to arrive at a solution containing uranium(IV) and vanadium(IV). It then electrolyzes the solution with an accurately controlled current, oxidizing the uranium to uranium(VI) and converting the vanadium to vanadium(V) at the electrode surface.

As it happens, vanadium(V) cannot coexist in the solution with uranium(IV). Thus the vanadium(V) that is formed immediately reverts to vanadium(IV) by converting uranium(IV) to uranium(VI). Only when all the uranium(IV) is gone can the vanadium(V)

concentration begin to build up. This causes a sudden change in the potential of the solution, indicating the end of titration.

The titration and the detection of its end point are both handled electrically; i.e., by controlling a current and measuring a voltage change, respectively. These measurements are extremely precise. They are also easy to obtain in digital form acceptable for computer processing.

Similarly, we perform all weightings on an electronic balance. The operator's role is to transfer each sample to the balance pan, advance a dial to find its weight to the nearest gram, and enter the sample's identifying number at the keyboard. The electronic balance completes the weighing to 0.1 mg and reports the weight directly to the computer, where it is stored for use in later calculations.

After the aliquots have been weighed, the operator transfers them to the automatic sample changer, identifying each by number and also entering its position in the sample changer. This gives the computer all the information it needs; it then proceeds with the titration and prints out the assay results.

Fig. 4. Computer-automated uranium assay system on display before shipment to ERDA's New Brunswick Laboratory.

Table 2. Chemistry of the modified Davis-Gray assay for uranium

1. Add sulfamic acid to remove NO_2 formed from the NO_x in step 3 below.
2. Add Fe(II) in strong H_3PO_4 . Phosphoric acid (H_3PO_4) acts as a complexing agent that enables Fe(II) to reduce U(VI) to U(IV) by the reaction: $\text{U(VI)} + \text{Fe(II)} [\text{excess}] \rightarrow \text{U(IV)} + \text{Fe(III)} + \text{Fe(II)} [\text{excess}]$.
3. Add HNO_3 and Mo(VI) catalyst.
 $\text{Fe(II)} + \text{HNO}_3 + \text{Mo(VI)} \rightarrow \text{Fe(III)} + \text{NO}_x$.
4. Add diluent containing V(IV) in dilute H_2SO_4 .
5. Titrate with electrogenerated V(V) .
 $\text{V(IV)} \rightarrow \text{V(V)} + e^-$.
 $\text{V(V)} + \text{U(IV)} \rightarrow \text{U(VI)} + \text{V(IV)}$.

To guard against unforeseen changes in operating conditions that might introduce errors -- and as a safety measure -- we incorporated an extensive fault-monitoring network in the analyzer. This network watches for spills and power outages, constantly checks flows and pressures within the system, and monitors several characteristics of the titration itself. Standard solutions of known uranium content are interspersed among the unknowns; as these standards are assayed, the network's software compares the results with the known assay and computes the deviation. If any of these checks show something amiss, the system is programmed to sound an alarm, indicate and record the fault, and then shut down, hold, or carry out corrective actions, depending on the nature of the fault and the point in the analytical cycle at which it occurs. All this cross checking greatly increases the credibility of the analytical results, minimizes the chance of processing samples incorrectly, and permits completely unattended operation.

System Description

Figure 4 displays the entire uranium titration system. The PDP-8/e minicomputer, the electronic balance, the sample changer, and the beaker-position reader are all commercially available. Special software and electronic hardware were developed for handling the operations of this system, including a control panel/interface that permits manual manipulations and provides a graphic display of system status. The titration and data acquisition are controlled by new instrumentation⁷ that regulates the electrolysis current and measures the potentials during electrolysis.

The analyzer is based on an assay proced ure

developed by Davies and Gray⁸ of the Dounreay Experimental Reactor Establishment, Scotland, and extended by investigators^{9,10} at ERDA's New Brunswick, N.J., Laboratory. The chemistry of our modified procedure is outlined in Table 2. The first step is the addition of a solution of sulfamic acid to prevent a buildup of nitrite ions in the third step. An excess of iron(II) in strong phosphoric acid is then added, reducing the uranium to uranium(IV). In the third step, the analyzer adds nitric acid with a molybdenum(VI) catalyst to oxidize the excess iron(II). Finally, it adds a solution of vanadium(IV) in dilute sulfuric acid, which readies the solution for titration. The sequence of photos in Fig. 5 shows the changes in appearance of the solutions as they go through this process.

Figure 6 shows the titration cell station, together with some of the details of the sample changer. Each aliquot, in its separate 300-ml beaker, occupies a numbered compartment in one of the sample trays. The sample changer accommodates 11 of these four-compartment trays, for a maximum of 44 samples.

When a beaker arrives beneath the titration station, the position indicator reads the position number from a perforated card on the side of the compartment and reports this number to the computer. A pneumatic piston then raises the beaker into position. A transparent plastic collar around the titration station prevents any misaligned beaker from rising and crushing the electrodes.

As the beaker touches the elastomer seal at the upper flange, a gentle puff of air pressurizes it momentarily. This is to check whether the cell is sealed. Any chipped or otherwise defective beaker that

fails to seal is rejected without an assay, and the rejection is noted in the computer printout.

After titration, the beaker is again pressurized, forcing the spent solutions out through the waste tube. The electrodes are then rinsed with a distilled-water spray in preparation for the next assay. The pneumatic piston then retracts, the sample changer indexes to the next position, and the cycle repeats.

System Performance

This system analyzes samples at the rate of about

four to seven per hour. Preliminary results with uranium-metal standards, U_3O_8 , and UO_2 indicate that system precision is about 0.03 mg in the range 12 to 180 mg of uranium. In the optimum sample-size range, 80 to 120 mg, the absolute accuracy, based on electrical calibrations of the instrumentation, is within about 0.1%.

The data printout summarizes all information gathered during the assay as well as the analysis results. It includes the sample weight, the sample solution weight, the aliquot weight, the weight of uranium in

Fig. 5. Steps in the processing of a uranium solution during the automatic assay. (a) The original yellowish U(VI) aliquot, in place but still clear of the electrode and other cell components. (b) The same aliquot after reduction with Fe(II) in strong H_3PO_4 . (c) Aliquot during oxidation with HNO_3 to remove the excess Fe(II); oxides of nitrogen formed in this step are removed by the sulfamic acid added earlier. (d) Oxidation is complete; all the iron is now Fe(III). (e) Vanadium(IV) in H_2SO_4 has been added to raise the level above the gold screen electrode; coulometric titration is in progress. (f) Titration is complete and the solutions have been blown over into the waste distribution system. The cell components are now being rinsed with distilled water in preparation for the next assay.

Fig. E. Titration station, reagent distribution valve unit, and sample changer. Each aliquot is in a separate 300-ml beaker in the four-compartment trays on the sample changer table. The beaker in position 1 is about to be raised into the titration station.

The position indicator (lower right) reads the punched card and transmits the number to the computer memory. The computer recalls the previously stored association between sample number and position number to correlate the titration information with the previous weighings.

After each assay, the sample tray indexes one position to the right. When all four samples in a tray have been assayed, the tray indexes toward the rear of the table. The sample changer accommodates up to 11 trays, for a maximum of 44 samples.

The large white box at the upper left contains the valve that control the flow of reagents, distilled water, waste solutions, and air. Tubes from this distribution system can be seen leading to the main flange. Also passing through hermetic seals in the flange are wires leading from the various electrodes to the electrical distribution panel at the upper right. At the top is the motor that agitates the solution with an impeller.

the aliquot, and the concentration of uranium in the original sample. It also includes values of the electrode potentials at various stages of the titration — useful information for diagnostic postmortems.

The Davies-Gray assay on which this analyzer is based is the most versatile assay method known for uranium. It applies to nuclear materials from all stages of the fuel cycle, and to most of them without any

separation or pretreatment. We expect that our analyzer — now being evaluated at ERDA's New Brunswick laboratory — will possess this same versatility along with the virtues of automation: economy, speed, and accuracy.

Key Words: titration; uranium — analysis; uranium — assay.

The slapper detonator, a radical departure from conventional exploding bridgewire detonators, is one of the developments of LLL research into detonator design parameters. Called the slapper because it uses a high-velocity impact to initiate detonation in high-density explosives, it has applications in nonnuclear ordnance and, potentially, in commercial blasting. It is compact, inexpensive, easy to make, resistant to environmental extremes, and even safer than the conventional exploding bridgewire detonator.

In the interests of safety, it is desirable to make high explosives as insensitive as possible. TNT, a commonly used explosive, can be struck with a hammer or set afire without detonating. It takes a particularly sharp shock to make it detonate. The function of the detonator is to provide this sharp shock at the desired time. The more energetic the detonator, the more insensitive the explosives can be. Insensitive explosives are not only safer to handle but also far less susceptible to homemade detonation schemes.

There are many varieties of detonators, including the simple percussion detonators found in shotgun, rifle, and pistol shells and the more sophisticated electrically fired blasting caps. These contain lead azide, a primary explosive, and can be set off by heat, shock, or electrical currents of only a few milliamperes. The exploding bridgewire detonator, a more expensive but much safer detonator, contains only secondary explosives and can be initiated only by a large pulse of electrical current.

The exploding bridgewire detonator (Fig. 7) consists of a fine wire imbedded in a low-density compact of a secondary explosive. To fire it, a large enough current pulse must be passed through the wire to make it explode. The energy of this explosion ignites the low-density secondary explosive compact; the shock of that detonation sets off the high-density booster that detonates the main charge.

This somewhat roundabout system is subject to several uncertainties and possible safety problems. The bridgewire must be made to exacting tolerances: it must have just the right combination of resistance and current-carrying capacity, it must be thin enough to explode but strong enough to withstand abuse without breaking, and it must be immune to chemical attack by the explosive in which it is imbedded. Conversely,

Fig. 7. Basic features of the exploding bridgewire and slapper detonators. As shown, the slapper detonator requires no low-density explosive, and the exploding foil bridge is completely separated from the high-density explosive, providing greater safety.

it must not change the explosive in any way that would affect its sensitivity.

The slapper design (Fig. 7) eliminates most of these worries. It operates by exploding a thin metal foil that drives an insulating film across a gap to set off the booster pellet directly by high-velocity impact. It eliminates the low-density compact of secondary explosive, and completely separates the exploding

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Fig. 8. Design configuration of the slapper, without the barrel and explosive. The color photo shows part of a sheet of metal-foil bridge elements.

Fig. 9. Construction of a complete slapper detonator. From top to bottom: coaxial cable is soldered to the electrodes of the substrate-bridge-flyer sandwich; a barrel made of plastic shim stock with a punched hole is bonded to the flyer; the explosive pellet is placed on top of the barrel, over the hole.

Fig. 10. Slapper detonator design parameters studied, including bridge, flyer, barrel, explosive, and other variables.

Fig. 11. Two prototype slapper assemblies: (left) an integrated detonator/cable assembly; the bridge element is etched on the end of the flat cable; (right) a ceramic/glass slapper detonator designed to withstand high temperatures.

metal foil from the explosive. This provides one of the slapper's greatest advantages - improved safety.

Slapper Design

Basically, the slapper is a printed-circuit assembly (Fig. 8) etched from a metal film on a laminated dielectric substrate. The substrate laminate consists of an acrylic layer, an aluminum back conductor, and two polyester-film insulators. The etched bridge on the top insulator may be of almost any metal, usually aluminum or copper.

Figure 9 shows the steps for building the complete detonator. First (top), coaxial cable is soldered to the electrodes of the substrate-bridge-flyer sandwich. Next (middle), a barrel made of plastic shim stock with a punched hole is bonded to the flyer. Then (bottom), the explosive pellet is bonded on top of the barrel, over the hole.

The design parameters we studied to develop the slapper are shown in Fig. 10. We considered most of the common metals as bridge materials. Copper and aluminum are both very good candidates because of their electrical burst behavior and their ease of fabrication.

For the flyer, we investigated glass, ceramic, most of the common plastics, and sandwiches of dielectrics and metals. We favor polyester and polyimide films because they perform well and are readily available in low-cost laminates.

The simplest barrel is a sheet of plastic shim stock with a punched hole. We studied the importance of hole size and precision and found that the dimensions are not particularly critical. The barrel is not really necessary, although it improves performance by reducing the detonating-power threshold.

We examined many of the common secondary explosives and found that most can be detonated with the slapper, given enough power and a large enough flyer. Our experiments show that HNS-I (a crystal form of 2,2',4,4',6,6'-hexanitrostilbene) works well with the slapper. It has a reasonably low detonating threshold and an initiation-failure dimension (minimum flyer diameter) of less than 1 mm. These properties permit design of a small detonator with a modest power supply.

HNS-I has poor mechanical strength. We found that adding a plastic binder allows automated fabrication of strong, well-formed explosive pellets. A special plastic-bonded material, referred to as RX28AS, was developed for this purpose.

We also considered placing a thin skin on the

explosive pellet to improve its mechanical integrity in high-shock environments. This can be done with very little effect on the power threshold.

The main criterion in our choice of a substrate material was ease of fabrication by automatic methods. Different substrate materials affect detonator behavior but the various effects are unimportant compared with this main criterion of fabrication ease.

We developed and fabricated prototype slapper assemblies for various practical applications. The left side of Fig. 11 shows a bridge element etched on the end of a flat cable, designed to minimize detonator thickness. This assembly is made of a polyimide film with copper conductors on both sides.

A slapper designed to withstand high temperatures appears on the right side of Fig. 11. It has a circuit of gold-glass ink deposited on an alumina substrate by an inexpensive silk-screening process and fused in place. The flyer is glass, and the barrel is alumina. This detonator's high-temperature performance is limited only by the temperature behavior of the explosive.

Advantages and Disadvantages

Safety is perhaps the slapper's most important advantage. The slapper has a detonating threshold even higher than that of exploding bridgewire assemblies (Table 3), and it uses rather insensitive secondary

Table 3. Typical detonating thresholds

Detonator	Voltage, V	Current, A	Energy, J
Exploding bridgewire	900	250	0.25
Slapper	3000	3000	2.5

explosives. These insensitive explosives are safer to handle and make detonation far more difficult for persons not equipped with suitable firing electronics.

To take advantage of the inherent safety of the slapper detonator, we used microcomputers to replace mechanical safing elements now used in conventional fuzes in nonnuclear ordnance. The prototype systems we built and demonstrated were very small and inexpensive. Such a system would be fast-firing, completely testable, immune to noise, and virtually fail-safe. The time it takes to charge the firing circuit capacitors is an added safety feature, practically precluding the possibility of accidental detonation.

Slappers are also attractive because of their resistance to environmental extremes. We have shown that they can stand high shock (approximately 2000 km/s²). They can be made to resist more heat than any explosive. Their performance is virtually unaffected by cold. Moisture in the explosive causes almost no problems, and we predict that long-term storage will be much less difficult than for exploding bridgewire detonators.

The low cost of making slappers is another advantage. Although we have not demonstrated automatic production, design and materials studies convince us that slappers can be mass-produced very inexpensively. We also find that the production controls necessary in making reliable slapper detonators are less stringent than those required for exploding bridgewire detonators.

A major design problem of the slapper is the size of the power supply needed to provide enough energy to explode the foil and detonate the booster. We made a considerable effort to minimize the firing circuit and achieved designs for complete firing systems with minimum weight and volume using stock components. The smallest weighs only 370 g. Further reductions are certainly possible using special components.

Overall, the advantages of the slapper detonator far outweigh the disadvantages. The safety implications, to both the blasting industry and the general public, warrant serious consideration.

Key Words: detonators; detonators - fabrication; exploding wires; explosive initiators; initiators; slapper detonators.

NEUTRON AND GAMMA-RAY TRANSPORT EXPERIMENTS IN LIQUID AIR

Accurate estimates of neutron and gamma radiations from a nuclear explosion and their subsequent transport through the atmosphere are vital to nuclear-weapon employment studies: i.e., for determining safety radii for aircraft crews, casualty and collateral-damage risk radii for tactical weapons, and the kill range from a high-yield defensive burst for a maneuvering reentry vehicle.

Radiation transport codes, such as the Laboratory's TARTNP, are used to calculate neutron and gamma fluences. We have been conducting experiments to check and update these codes. Recently, we used a 1.3-m-radius liquid-air (21% oxygen) sphere, with a pulsed source of 14-MeV neutrons at its center, to measure the fluence and spectra of emerging neutrons and secondary gamma rays. Comparison of measured radiation dose with TARTNP showed agreement within 10%.

Neutrons emitted from nuclear explosions interact in the surrounding air and produce secondary gamma rays. Depending on their intensities and the distance from the source, these radiations may kill exposed individuals, irradiate physical structures, or pose a hazard to friendly troops and civilians. Accurate estimates of their intensity after transport through various ranges in air thus are vital data.

Radiation transport codes are needed to calculate neutron and gamma fluences because measurements of neutron transport through long ranges in the atmosphere are not feasible without highly intense sources like nuclear explosions. Such calculations are usually reliable up to a few mean free paths. For larger distances, however, any coding errors and approximations or cross-section uncertainties can give large errors in the calculated fluences. Recent LLL benchmark experiments - involving a sphere of liquid air with a pulsed source of 14-MeV neutrons at its center and neutron/photon detectors at a distance from the sphere - have provided an excellent means of checking the accuracy of our codes.

In the past, LLL measurements of neutron fluences from pulsed liquid-nitrogen spheres of 11-, 19- and 56-cm and 1.3-m radii have given results^{11,12} in good agreement with code calculations. Experiments elsewhere¹³ involving large volumes of liquid nitrogen have provided time-integrated data, but their agreement with code calculations is not satisfactory for all energies or for long distances.

Our experiments with the 1.3-m-radius liquid-air sphere had a simple geometry and a low-background detector setup that obtained differential data by the neutron time-of-flight technique. A liquid-air sphere of this size is roughly equivalent to the amount of air in a 1-km sphere of atmosphere. Although our results are not similar to those that would be obtained under

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Fig. 12. Experimental setup for pulsed-sphere experiments. The 10-m³ liquid-air sphere surrounds a 20-mm-diam D-T neutron target. The sphere was a 2.6-m-i.d. Dewar flask with a 5.3-mm-thick copper-alloy shell and a 4.7-mm-thick steel outer vacuum enclosure (3.3-m o.d.). A liquid-scintillator detector at 26° with respect to the deuteron beam is placed 9.5 m from the source in a low-background room. A second liquid-scintillator detector is placed 9.6 m away at 120° to the beam, in a deep hole in the ICT pit wall. Measurements from the 26° detector are regarded as more reliable because it views a better-defined volume of the sphere.

actual atmospheric conditions, the experiment provides a stringent check on fluence and dose calculations by the TARTNP code up to 1 Mg/m² of liquid air and gives confidence in calculations under actual atmospheric conditions for even longer ranges. The experimental results are also consistent with dose

measurements made simultaneously with a tissue-equivalent ion chamber.

Because there are many uncertainties associated with weapon applications, considerable uncertainty is permissible in estimates of neutron and gamma radiation. Weapon delivery and effects systematics

suggest that a reasonable goal for the uncertainty in calculated dose is +50%, -33% at any range. In our latest work, we have made another step toward that goal.

Experimental Setup

The measurements were made with the LLL Insulated-Core-Transformer (ICT) accelerator as a pulsed source of 400-keV D^+ ions, producing neutrons. Figure 12 shows the experimental setup. The deuteron beam was steered to a 20-mm-diam, tritium-loaded titanium target at the center of the liquid-air sphere.

The oxygen/nitrogen ratio of the liquid air was monitored continuously during the runs and maintained at a value of $21 \pm 1\%$ oxygen. Since nitrogen has a lower boiling point than oxygen, it boils off more rapidly, leaving an oxygen-rich mixture. To avoid this, liquid nitrogen was added periodically. When the oxygen ratio dropped, oxygen-rich liquid air was added. The additions to the Dewar and some internal boiling kept the elements mixed, which was recorded by the mixture monitor.

Neutron and photon detectors were located at two different positions outside the sphere to obtain two independent sets of data for comparison with code calculations. High-energy neutrons and gamma rays were measured in separate runs. A liquid-scintillator detector was placed at 26° with respect to the deuteron beam and viewed only the liquid-air volume and the inner shell of the vessel. To measure low-energy neutrons, we used a 6Li -loaded glass scintillator detector in its place. A second liquid-scintillator detector, placed at 120° to the beam, viewed a cone-shaped volume of liquid air whose 1-m base was centered at the liquid-air center with its apex at the detector. The data from this detector are less reliable because it viewed a less well-defined portion of the total volume of liquid air.

We recorded the signals from the detectors and obtained time-of-arrival and pulse-heights information about the radiations emerging from the liquid-air sphere. These data were later processed for comparison with calculations.

Data Collection and Analysis

High-Energy Neutrons. To obtain high-energy neutron data, we operated the ICT accelerator with a pulse width of 4 ns and a pulse rate of 500 kHz. The differential measurements to separate neutrons of various energies emerging from the liquid-air sphere were made by observing their time of arrival at the

liquid-scintillator detector relative to the time of the neutron-producing beam burst at the center of the sphere. Of all the pulses made by the neutrons arriving at the detector, those produced by recoiling protons had pulse heights well above the noise and were the principal component involved in neutron detection. We used pulse-shape discrimination to suppress pulses produced by gamma rays during this run.

Because the uncertainty of a detected neutron's origin is not negligible compared to the source-to-detector distance, the relation between energy and time of arrival has a precise meaning only for uncollided neutrons and for those transmitted after interactions near the source. However, a comparison of the measured time-of-arrival data with TARTNP calculations for the simulated experiment is strictly valid because all the conditions affecting the data and all materials, in addition to the liquid air, are included in the calculations.

From the experimental time-of-arrival data for the liquid-air run, we obtained a spectrum expressing the neutron time of arrival as a function of neutron count rate divided by the expected total source neutron count during a blank run (Dewar without liquid air). To obtain this total source neutron count, we compared the monitor alpha counts during the liquid-air run with the monitor alpha counts during a blank run. This procedure avoids any error in determining detector efficiency; it is subject only to the error in estimating efficiency at 14 MeV relative to that at all other energies.

A special edit of the TARTNP calculation gives space, time, and energy coordinates for any neutron entering a selected zone; for our calculation, we chose a zone extending from 14° to 26° with respect to the deuteron beam. Figure 13 compares this calculation, with the detector efficiency and overall resolution folded in, to the 26° experimental data. The total integral counts are very close, but about 13% fewer neutron counts at early times of arrival (8- to 15-MeV equivalent) are observed in the calculation compared with those in the measurements.

Results for the 120° detector give qualitative agreement and are also shown in Fig. 13. However, because the liquid-air volume viewed by the detector is somewhat uncertain, we shall concentrate on the 26° data for numerical comparisons.

Low-Energy Neutrons. We measured the spectrum of low-energy neutrons emerging from the liquid-air sphere by replacing the 26° liquid-scintillator detector with the 6Li -glass detector. We used two beam pulse

Fig. 13. Time-of-arrival spectra for high-energy neutrons measured with 26° detector (top) and 120° detector (bottom), both compared to TARTNP calculations.

rates — 1 and 10 kHz — to obtain a correct background for both early and late times. The agreement between the experimental and the calculated neutron counts is good if we use the

early-time data up to 30 μ s from the 10-kHz run and the late-time data from the 1-kHz run (Fig. 14). The low-energy neutrons contribute roughly 25% of the dose and can be important in

checking the code for low-energy neutron transport.

Gamma Rays. To measure the gamma fluence, we calibrated the scintillation detectors using gamma sources and set the lower pulse-height bias level at half the Compton edge of the 511-keV gamma-ray spectrum. The gamma rays due to noncapture neutron interaction in liquid air were separated quite well from

neutrons arriving at the detector because of their time of flight. We measured a pulse-height spectrum of all gamma rays occurring within a 60-ns time window following a beam burst.

We then compared the gamma-ray-produced electron pulse-height spectrum measured with the 26° scintillator with the results of a TARTNP calculation after folding in the detector response for gamma rays

Fig. 14. Time-of-arrival spectra for low-energy neutrons measured with the ^6Li -glass detector compared to TARTNP calculations: (top) data from the 10-kHz run; (bottom) data from the 1-kHz run.

Fig. 15. Gamma-ray pulse-height spectrum measured with the 26° detector, compared with TARTNP calculations.

(Fig. 15). The integral of the measured data for this detector is only 2% lower than the calculations, but the calculated data have a preponderance of high-energy electron pulses. Possible sources of error are the calculated gamma production and attenuation, the calculated detector efficiency, and experimental errors. Because of the complicated calculational geometry involved, we have not yet done the gamma-ray calculation for the 120° detector.

Radiation Doses from Neutrons and Gamma Rays.

A rough estimate of radiation dose levels in tissue resulting from neutrons can be obtained if we multiply the TARTNP output by published kerma factors (conversion factors from fluence to dose).¹⁴ Multiplying TARTNP fluence groups by the appropriate kerma factors and summing the products, we obtained the calculated total neutron dose down to a few electron volts. The value is 30.1 aGy (3.01×10^{-17} rads) per source neutron at the detector location.

We have also derived the experimental integrated dose per source neutron at the detector location (by comparing the measured fluence in various time intervals with the calculated fluence) and obtained an approximate value 33.0 aGy (3.30×10^{-17} rads); this is about 10% higher than the calculated value. We used the experimental and calculated gamma-ray pulse-height spectra to derive directly the expected doses at the detector location.

Discussion

Our integrated neutron and gamma fluences and estimated doses are presented, for a qualitative comparison of measurements and calculations, in Table 4; the differential data are represented in the figures. The statistical uncertainties in the differential data range from 7 to 15% at each point, but the integral-data statistical errors are much smaller. The main sources of systematic error result from the detector efficiencies that have been folded into the calculations and dominate other errors. A systematic error of about 7% may be assumed in the high-energy neutron-detection efficiency and in the gamma detector efficiency, whereas the ⁶Li-glass-detector efficiency has an uncertainty of 15 to 20%. Since the measured errors are mostly statistical and the systematic errors are all transferred to the calculations, the error in the total calculated integral dose is the sum of the error in the calculations and their discrepancy with measurements. We therefore expect the dose calculations to be reliable to 15%.

Although the experimental dose resulting from the neutron spectrum is only about 10% higher than the calculations, the 13% difference in neutron counts above 8 MeV is worth investigating. Possibly more forward elastic scattering or a change in inelastic cross sections could provide better agreement, if incorporated in the code. A decrease in nonelastic cross sections could also yield a lower number of calculated

Table 4. Summary of integral results for neutron and gamma rays

	Experiment	Calculation	Ratio
Neutron results			
Integral neutron count rate (2 to 15 MeV), counts/ns/blank-run counts	5.81×10^{-2}	5.81×10^{-2}	1.00
Integral neutron count rate (110 eV to 1 MeV), counts/ns/source neutron	3.58×10^{-9}	3.43×10^{-9}	1.05
Estimated integral neutron dose/source neutron, aGy ^a	33.0	30.1	1.10
Gamma-ray results			
Integral gamma-ray count/source neutron (early-time window only)	2.64×10^{-8}	2.70×10^{-8}	0.98
Estimated integral gamma dose/source neutron (early-time window only), aGy ^a	8.4	9.2	0.91
Combined results			
Neutron-plus-gamma dose/source neutron, aGy ^a	41.4	39.3	1.05

^aaGy (attograys) = 10^{-18} gray = 10^{-16} rads.

gamma rays. The agreement between calculated and measured gamma rays is reasonably good.

Conclusions

This work verifies our knowledge from earlier work on neutron and gamma-ray transport from a nuclear burst and provides a benchmark on the dose-vs-range curve. For 1 Mg/m² of air, the calculations yield integral doses that have an accuracy of about 15%. The radiation doses calculated with the TARTNP code for 1 km of atmospheric air could therefore be accurate to 15%. The extrapolation of these calculations to longer ranges can be done with a fair

degree of confidence. At longer ranges, we expect that elastic scattering will predominately determine the neutron transport and that the gamma fluences will be determined largely by attenuation of gamma rays already produced near the source. Production due to capture doses does not become important until much longer ranges or for lower energy neutron sources.

These data provide an excellent validation of the cross sections as well as the codes.

Key Words: pulsed sphere measurements; integral tissue dose; neutron transport; gamma ray transport; radiation transport; radiation transport computations; TARTNP; pulsed liquid-air sphere.

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