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A SUMMARY OF BATTELLE-NORTHWEST
LABORATORY CONTRIBUTIONS TO MFC-12

E. A. Eschbach
W. A. Reardon

September 1970

AEC RESEARCH &
DEVELOPMENT REPORT

BNWL-1423

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TABLE OF CONTENTS

	<u>Page</u>
I. INTRODUCTION	1
II. TESTING SERVICES	5
III. ISOTOPE PRODUCTION	10
IV. POTENTIAL PRODUCTION OF LOW-SHIELDING-TYPE RADIOISOTOPIC HEAT SOURCES .	15
V. SUMMARY OF FLUX TRAPPING	32
APPENDIX A: RAMPARTS AND NOVEL CONTAINMENT	A-1

LIST OF TABLES

<u>Table Number</u>	<u>Title</u>	<u>Page</u>
I	Effective Absorption Cross Sections Versus Neutron Temperature And Spectrum Hardness For Selected Isotopes	14
II	Characteristics of Radioisotopic Heat Sources	19
III	Figure of Merit To Compare	25
IV	Effect of Trap Size And Driver Moderator Density On ϕ_T/ϕ_D	26
V	Effect of Trap Density On ϕ_T/ϕ_D	27
VI	Effect of U-235 In Trap On ϕ_T/ϕ_D	29

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I. INTRODUCTION

The major intent of MFC-12 was to conceive ways in which Hanford production reactors could be altered to increase their versatility in nuclear products and services by changing moderator-to-fuel ratio and perhaps the moderator. In undertaking this effort, it was recognized that a wide variety of markets must exist, and that much effort would be required to identify those most scientifically and economically viable. In this world of rapidly changing technology and demands, some of the prospects that were waxing at the time of study have now waned. Nevertheless, we shall report these prospects simply because what was promising two years ago and is unpromising today may be promising tomorrow.

In observing the characteristics of the old Hanford reactors, it was found that several major characteristics appeared to be relevant to the considerations of MFC-12, to wit:

1. Large integral neutron flux volume.
2. Charging-face-accessibility 100 percent of the time, even during operation.
3. Non-recirculating coolant. (This has a very handy feature beyond front-face (charging face) accessibility throughout operation, namely, the pumps and valving of cooling system cannot become contaminated. However, it poses a problem of contamination of a heat sink, although with adequate damming and diverting devices, this may be circumvented.)

4. Solid moderator, that is, graphite. (This has advantages from the stand-point of supporting the process tubes, etc., mechanically. It has the disadvantage, perhaps only incidental, of making it difficult to change the ratio.)
5. Process tubes rather than a large kettle. (This allows individual piping to each tube, and thus fuel entity, adjustment of the flow, specific instrumentation, etc., and on balance is an asset for a production reactor.)
6. No power generation. (There is no by-product. The cost of fission must be borne by the products made. However, without power generation, there is less incentive to operate with bulk-coolant temperatures above the atmospheric boiling point of water. Thus little pressure-volume energy must be accommodated by a containment system.)
7. No reactor containment. (This may pose a problem on starting up a deactivated unit.)
8. Hanford is a complete nuclear site. The site includes a very large processing complex equipped with considerable electrical and water supply systems, as well as basins, outlet pipes, and the like. (Such a site may well be optimum for a device that can be used as an adjunct to a reactor, such as an accelerator.)

The possibility of altering the moderator-to-fuel ratio, or even the type of moderator within the reactor, led us to reconsider the cooling and containment systems and other pertinent adjuncts to the reactor. For this reason, before we proceeded very far with analyzing the reactor alterations proper, we did considerable analyses of whether or not more than just the reactor proper should be changed, that is: Should we recirculate the coolant? Should we generate some electrical power? Should we use vertical rather than front-to-rear tubing holes? Should we make provisions for reactor containment? These questions arose because it was recognized that if any of them were given affirmative answers in the future, the changes involved might be blocked if the moderator-to-fuel ratio, or the moderator itself,

were changed. Although providing answers to those questions was not the major objective of the study, the answers arrived at are mentioned here because they may be important both for other studies and for the applications at hand. After considerable analysis, we concluded:

1. We should not recirculate the coolant. (The cost associated with the recirculating system appears uneconomic except for those cases in which it is mandatory, i.e. D_2O or Na.) Not only is the capital cost large, but it sacrifices too much of the existing reactors capability. (See 2 below.)
2. It appears that we should maintain front-face accessibility. This is an asset to the system and, if compromised, would seriously hamper low-cost operations.
3. We should use front-to-rear rather than vertical holes. Although this may preclude certain applications that require criticality, it appears to make little or no difference for most applications. And the cost of maintaining front-to-rear holes is so low in the existing reactors as to make their use almost mandatory. This does not block, however, the possibility of a few vertical holes, especially if these do not have large diameters.
4. If containment is required, as in the startup of a presently shutdown reactor, it is almost mandatory that some novel scheme of containment and operating techniques be employed in order to be feasible at all and to control costs. How this might be done is explored in Appendix A.
5. Power generation should not be considered within the framework of MFC-12, the principal reasons being the lack of demand, the long pay-off period, and the complications resulting from dual purpose operation.

Within the above limitations, the two major reactor alterations considered in the MFC-12 Program were the following:

1. To supply extensive testing capability for both fuels and other materials in both thermal and fast neutron fluxes.
2. To provide special holes or regions in the reactor for irradiating a variety of targets.

The market to be affected by either of these alterations is uncertain at this time. However, the testing capabilities and versatile irradiation facilities appear unique and will be discussed separately.

II. TESTING SERVICES

Testing services cover a multitude of activities, but those considered in this study relate to the load processes for which Hanford reactors would have a distinct and unique capability. Since fuels testing is being performed in other areas besides Hanford, and since commercial reactors are doing their own in-situ fuels testing, we did not think ourselves competitive in full-testing applications, and so did not delve into this field.

However, materials testing for radiation-damage studies offers a field wherein the Hanford reactors can serve as a unique instrument. It was determined early in the studies (see DUN-AOP-6. October 1966) that the excavation of holes for large space-type irradiations seemed feasible, the holes being as large as six feet in diameter and extending horizontally from the front face of the reactors. Such large-space-type irradiations would indeed make the Hanford reactors unique as test instruments. The kind of irradiation that might be accommodated in such large spaces would make it possible to study electronic devices in such a way that an entire device could be inserted in the reactor and its operation checked as a function of the neutron irradiation. The damage to be expected from neutron irradiations would be caused by changes in crystal structures, the tensile strength of relay arms, and the like.

Thermal neutron fluxes of the order of 10^{12} nv are probably completely adequate for testing in such large spaces. It is also probable that only minimum cooling would be required, and that this requirement would be due mainly to n-gamma reactions and absorption of incident gamma rays. The cooling for this sort of a testing can probably be handled adequately by air, helium, or other gases. A very high temperature space can probably be achieved either by throttling the coolant or by inserting additional heaters. In addition, some spectral adjustment is probably available from in the manner which the cavity is cooled or the manner in which it is interfaced with the remainder of the reactor.

If it is desired to increase the fluxes beyond the prevailing levels of a normal operating reactor, then perhaps recourse can be had to flux trapping or other methods of enhancement of the flux. The flux trapping is equally

applicable to the special holes which will be discussed later in Section III of this report. (See page 10.)

The market potential for large irradiation spaces that could be uniquely provided by the Hanford reactors was examined and reported in DUN-AOP-23.⁽¹⁾ Evaluating pressure-vessel steels is one of the most promising prospects because the fractural mechanics studies in such evaluation work would require very large samples and an unclassified fast flux, namely, one with energy greater than 0.1 MeV and on the order of 5×10^{12} nv. Fractural mechanics studies would also require an isotropic flux in a volume whose dimensions are 12 inches in diameter by 40 inches long. Some material testing requires even larger spaces. Evaluating process-tube materials, for example, would require both a flux of about 10^{13} nv and reactor holes of about 4 inches in diameter. Possibly, the simplest approach to achieving a high fast flux in the reactor hole would be to simply provide a peripheral converter plate. If one were to use a converter plate, the fluxes attainable would be two or three times greater than those presently achievable. Of course, the cooling requirements would be relatively severe because of the fissions in the converter plate.

In most of our studies when the test cavities were not permitted to have individual multiplication, the fluxes were too low to be of great interest. A low flux would also occur with a converter plate as well as with any of the concepts calling for a driver zone. Because of this problem of low flux, several studies were undertaken in which the fast flux module would be allowed, within limits, to be independently critical. The concepts which were examined were basically the gas-cooled concept, the sodium-cooled concept, and the **steam-cooled** concept. These three types of facilities will be discussed in order:

a) The Gas-Cooled Concept. A study to evaluate the feasibility of locating a fast flux cavity in a Hanford reactor was undertaken by G. F. Bailey (See "Fast Flux Cavity Capability" report by G. F. Bailey, DUN-2184, March 1967). This reference design consisted of a 4-inch-diameter test channel surrounded by a 10-inch-thick annular driver zone fueled with Inconel-clad uranium dioxide

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(1) DUN-AOP-23, MONTHLY REPORT, November-December 1967, p.7 (Secret).

dispersed in metal plates. The surrounding buffer zone consisted of 3 inches of depleted uranium at about one-half the metal density. Although preliminary calculations suggested that fast fluxes of about several times the presently available thermal fluxes might be obtained, a subsequent and more detailed analysis indicated that fluxes only on the order of the available thermal fluxes were probably obtainable. A subsequent study⁽¹⁾ examined a few possible variations by which one might enhance the fast flux over the surrounding reactor thermal flux. In one variation a 1-1/2 fold increase could be obtained if one creates a flux trap on the outside of the buffer which surrounded the fission plate. This method has some merit because the coolant is D₂O in the buffer and the fission plate, rather than a gas, and is somewhat more effective. The cavity itself, however, remains gas-cooled. Both of the above studies also indicated that the most promising concepts were the ones wherein the test cavity would be permitted to be independently critical.

In another variation, the fast-neutron multiplying region could be de-coupled from the surrounding reactor core by a thermalizing graphite buffer and some thermal neutron absorber, the absorbing region being used for some special isotope production. It is reasonably clear, however, that the mixture of these two kinds of reactors will present some control problems unless they are very thoroughly de-coupled. However, taken independently, it may be that one would not necessarily wish to fuel the rest of the reactor anyway. The gas-cooled concept presented a large number of problems and was temporarily abandoned when it appeared that the sodium-cooled loops would be of most interest.

b) Sodium-Cooled Concept. The sodium-cooled loops also presented problems of recirculating the sodium-coolant, transferring the heat, and the generally messy problem of inserting such a loop into an existing reactor. This concept was abandoned because of the possible competitions with FFTF and other programs for which the sodium-cooled loops could probably not compete.

c) Steam-Cooled Concept. Perhaps the most successful of the studies for test space was that of the steam-cooled, fast-test module. A unique design for

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(1) BNWL-CC-1353, "Transport Theory Analysis of Proposed Fast Flux Cavity In A Hanford Reactor", Richey and Humes, September 1957.

a fast-flux test module was evolved by combining a fast thermal system with a steam-cooled fast reactor. In this design, each module provides a fast flux of about 2×10^{15} nv, in a region 4 to 5 inches in diameter and 3 to 4 feet in length. The total core size of each module is about 30 inches in diameter: and this core is, in turn, surrounded by a neutron moderating reflector such as graphite, which would yield attendant thermal fluxes of about 10^{15} nv.

An important feature of this steam-cooled, fast-test module is the inherent fail-safe design. Calculations indicate that a module design can have maximum reactivity near the operating steam density, and that there will be (1) a negative change of reactivity upon loss of coolant due to the increased fast neutron leakage, and (2) a negative change in reactivity upon water flooding due to the presence of a thermal neutron absorber. By adding gadolinium oxide to the fuel, it is possible to ensure that the reactor will always be sub-critical when flooded with water. This safety feature also simplifies the mechanisms for discharging and handling the irradiated fuel, and allows the use of standard water-basin systems. Thus, although the steam-cooled fast reactors are not being considered for the power breeder reactors, the use of steam cooling for a test reactor is still very attractive.

When the steam-cooled, fast-test module is surrounded by a thermal neutron reflector, some further advantages are obtained. First, the thermal neutrons, by causing fission in the band around the outer edge of the module, create a driver region for the module. And this thermal driver tends to make the operating characteristics, namely, the neutron lifetime and the reactor-control characteristics of the module, more nearly that of the thermal reactor. The dependence on the thermal region makes a decrease in reactivity upon compaction that might occur, for example, by core melting. Second, the design combination of the size of the module and the pressure-vessel thickness can be used to give a nearly flat radial power distribution in a smooth transition between the fissioning in the thermal neutron driver to that in the fast neutron core. Finally, the thermal neutron flux peaking in the graphite a few inches outside the module, provides a large region of intense thermal neutron flux (about 10^{15} nv). This flux could be used for producing isotopes or for thermal neutron research. Combinations of the test modules surrounding a thermal neutron trap might be used to drive the flux intensity

even higher, and provide several fast test regions in the same reactor. It is also relevant that the steam-cooled module need not use a recirculating coolant, and that front-face accessibility is preserved as well.

III. ISOTOPE PRODUCTION

In order to produce competitively valuable isotopes in the Hanford reactors, one has several things to consider: first, the naive form of the reaction rate, (R), which is given by Equation (1) wherein

$$R = \phi N \Sigma V \quad (1)$$

where:

ϕ is the neutron flux

N is the atom density of the target nucleus

Σ is the production cross section

V is the volume.

While these four quantities do not vary independently of each other over a wide range, within a smaller range they certainly do, and some adjustment of them can be made. For instance, if the cross section Σ is small, one can increase the total reaction rate by increasing the flux, the nuclei density, or the volume, or all of them simultaneously. Fluxes, however, will generally be fixed by available power levels, and the atom density by available channels, so that volume is the final variable. The Hanford reactors, because of their very large volume, are in a good competitive position for producing isotopes, at least, those isotopes having a significant economic value. An example of this is the irradiation of bismuth to produce polonium. It is not true, however, that the flux cannot be altered, so one may take advantage of the so-called flux trap and boost the thermal fluxes to a higher level and consequently get a higher reaction rate. Each of the quantities in Equation (1), of course, has certain limits beyond which the physical capabilities of the system cannot go.

Still another way of looking at the production rate of a potentially valuable nucleus is given in Equation (2):

$$\frac{dN^i}{dt} = \sigma_a^{i-1} \phi N^{i-1} - (\sigma_a^i \phi + \lambda_i) N^i \quad (2)$$

On the left-hand side we have the rate of change of the i'th nucleus. On the right-hand side we have the formation rate given by $\sigma_a^{i-1} \phi N^{i-1}$, N^{i-1} being the atom density

of the target. Competing with this production rate, however, is the burnout and decay processes given by the second term $(\sigma_a^i \phi + \lambda_i) N_i^i$. The leading term, $\phi \Sigma_A$, is the ordinary burnout term whereas the $\lambda_i N_i^i$ would be the natural radioactive decay rate. These two parts of the second term may be regarded as competing processes insofar as they lead to quite different nuclei as an end-product. Equations (3) and (4) illustrate this difference in the way the two reactions behave:

$$\sigma_a^i \phi N_i^i(Z, A) \rightarrow N(Z, A + 1) \quad (3)$$

$$\lambda_i N_i^i(Z, A) \xrightarrow{\beta} N(Z + 1, A) \quad (4)$$

Equation (3) indicates how the burnup process leads to a nucleus with the same charge, but with a mass one unit larger. Equation (4) illustrates a beta-decay process in which the nucleus decays to a nucleus of the same mass, but with one unit higher charge. An illustration of the process of Equation (3) is the transmutation of Pu-239 to Pu-240. An illustration of Equation (4) is the decay process by which Am-244 becomes Cm-244. It is also interesting to observe that because of the competition between the two parts of the second term on the right-hand side of Equation (2) it is clear that the relative size of two parts of that term can be adjusted, depending on the relative size of $\Sigma_a \phi$, and the quantity λ_i . For instance, if it is desired to produce a large quantity of the material that comes from the beta-decay process, or the $\lambda_i N_i^i$, and if the competing cross section Σ_a is reasonably large, then it is advantageous to lower the flux because this will maximize the quantity N_i^i as a function of time. Nothing can be done about the λ_i , and one may see from Equation (5), which is the equilibrium ratio of the nucleus produced to the parent nucleus is given by the ratio of the production rate to the destruction and decay rate.

$$\frac{N_i^i}{N^{i-1}} = \frac{\phi \sigma_a^{i-1}}{(\sigma_a^i \phi + \lambda_i)} \quad (5)$$

We know from the simple radioactive decay law that the maximum of the isotope produced from N_i^i will depend on having N_i^i a maximum. Therefore, if we minimize

the denominator on the left-hand side of the equation, we will have a maximum of N^i . Conversely, if we wish to maximize the production of the quantity $N(Z,A+1)$, we will maximize the flux and, therefore, try to make this term dominate the natural decay rate. Another example may be found in the irradiation of Am-241, in which the bulk of the transmutation goes to Am-242, which has a 2500 barns fission cross section and a 16-hour half life. It is, therefore, advantageous to try to minimize the $\phi \Sigma_f$ as compared to the λ term in this isotope to produce Am-243. There are other examples of this competition in the various chains on the way to producing the higher transuranic elements.

There are still other variations available to the designer, namely, while ordinarily Σ is thought of as a property of the material and, in deed is, its effective values are also a function of several other parameters, one of the most important of which is neutron temperature. In the case of some nuclei having a low-lying resonance, such as Pu-239 whose resonance lies at about 0.3 eV is very large and has a very high capture-to-fission ratio, one can maximize the production of Pu-240 by raising the temperature and taking advantage of the high capture-to-fission ratio of this resonance. Still, another example is that of Np-237. This particular nucleus will be reported on in detail by E. T. Merrill in BNWL-CC-2000-AOPG. The essential feature, however, may be pointed out. The effective cross section at room temperature for Np-237 is 171 barns. If one hardens the spectrum by increasing the intermediate-to-thermal flux ratio, one can increase the cross section up to about 540 barns. One can accomplish the same thing, however, by maintaining a soft spectrum, i.e., high thermal-to-intermediate ratio, by merely increasing the temperature. If, in fact, one can raise the temperature to 2000°C, the effective cross section of Np-237 is about 630 barns. Such a cross section is a distinct advantage when irradiating Np-237 to produce Pu-238, because the principal contaminant of the Pu-238 is Pu-236, which is produced by n-2n reaction of Np-237. The n-2n reaction is basically proportional to the amount of fast flux available, or the power density. If one maintains a constant power density and increases the temperature, one may increase the reaction rate by about a factor of 4, and at the same time hold down the production of Pu-236.

The report, BNWL-CC-2000-AOPG, gives the details of the computer program written to tabulate isotopic cross sections as functions of neutron temperature,

spectra index, and fuel cell design. Each page of the report contains cross sections at 11 temperatures from 20°C to 3000°C and nine values of spectra index from 0.001 to 0.5. Three cell design parameters were included to cover such massive elements as those in the NPR, such small elements as those in the PWRs, and the homogeneous limit. Thus the 297 values could be considered as representative of any operating or proposed reactor, excluding fast reactors. Optional output from this program consists of normalized cross sections. The inputs selected consist of the temperature value, the spectra index, and the cell design parameter. This program also prepares a binary library for use of a one-step **burnup** routine.

The **burnup** routine will calculate the composition of a target sample of up to 16 isotopes. It prepares a table for each of the isotopes, for each of the 297 parameterized cross sections. Three types of tables are produced: (1) amount of isotope present at discharge, (2) grams of product produced per gram of target burned, and (3) product composition expressed as a percent of the product isotope in the product.

Table I gives an example of the change in effective cross section for three important isotopes: Pu-242, Np-237, and U-236.

TABLE I
EFFECTIVE ABSORPTION CROSS SECTIONS VERSUS NEUTRON TEMPERATURE
AND SPECTRUM HARDNESS FOR SELECTED ISOTOPES

Pu-242	<u>R⁽¹⁾</u>	<u>20°C</u>	<u>600°C</u>	<u>1000°C</u>	<u>2000°C</u>
	0.001	26.5	23.2	23.2	26.4
	0.100	37.3	47.2	54.5	80.5
	0.200	48.2	71.3	86.0	135.2
	0.300	59.1	95.5	117.6	190.0
	0.400	70.0	119.7	149.2	244.6
Np-237	0.001	170.5	216.5	332.6	629.7
	0.100	182.5	247.5	361.2	622.9
	0.200	194.6	278.8	390.1	615.9
	0.300	206.8	310.1	419.0	609.0
	0.400	218.9	341.4	447.9	602.0
U-236	0.001	6.2	5.0	4.8	4.7
	0.100	9.9	13.3	15.7	21.7
	0.200	13.7	21.7	26.7	38.8
	0.300	17.4	30.1	37.8	55.9
	0.400	21.2	38.5	48.8	73.1

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(1) R is the Wescott "r"; small r denotes soft spectrum and large R denotes hard spectrum.

IV. POTENTIAL PRODUCTION OF LOW-SHIELDING-TYPE RADIOISOTOPIC HEAT SOURCES

NASA has essentially "set the stage" for large-scale use of radioisotopic heat sources by making a request to the AEC for a minimum of 500 thermal kilowatts of Pu-238 by 1980. It appears that substantial amounts of this material could be produced by the AEC and by irradiating the neptunium recovered from spent, civilian nuclear power fuel. In addition, it is conceivable that a few other radioisotopes would be equally or better utilized than Pu-238. Thus, if these were available at equivalent cost, they could be substituted for, or used in addition to, Pu-238. The only isotopes which can be considered in this way include Pm-147, Tm-171, Tl-204, and Tm-170. These would be the most logical candidates since they require relatively low shielding and have reasonably long half-lives: two to four years. The following discussion examines the means of producing these isotopes (as a replacement or substitute for Pu-238) in amounts equivalent to 1000 kW of Pu-238.

If, by 1980, an artificial human heart using radioisotopic power is developed, Pu-238 would be in great demand. This use alone would require a minimum of 3000 kW per year (100,000 patients at 30 thermal watts each). Such a use obviously overshadows the "modest" NASA requirement. And since no prospects exist for such levels of Pu-238 production by 1980, other radioisotopes would have to be considered,

Promethium-147

This isotope is available today only via its existence as a fission product. By 1980, when the operating civilian nuclear power capacity in this country is about 110,000 MW_e, only 45.4 kW of useful (aged) Pm-147 can be expected. Its availability will, of course, increase in direct proportion to the increase in nuclear power. Even so, the available Pm-147 will be far less than the Pu-238 that could be obtained by irradiating neptunium which has been processed exclusively from spent, civilian power reactor fuels,

The only known means of increasing the supply of Pm-147 is to irradiate the neodymium obtained from mineral sources. Neodymium is not a rare element. It exists in the earth's crust at higher concentrations than those of such familiar elements as cobalt, lead, bromine, molybdenum, antimony, arsenic, boron, and uranium. Normal mineral sources (monazite and bastnesite) today could supply about 100 tons per year of neodymium from which, we can assume, it would be possible to recover 10 tons of pure Nd-146 by some yet unknown and economical isotope-separation technique. However, for a continuing program, this neodymium production could essentially double each year. Furthermore, if the need existed for greater outputs, impressive new sources of neodymium could be developed to increase production by at least a factor of 10. The U.S. wet phosphoric-acid industry alone could conceivably recover 1000 tons of neodymium annually. Neodymium as a by-product from the expanding europium industry (for color television) would also be an additional impressive source. By early use of such resources, combined with recycle of Nd-146, it is conceivable that Pm-147 production approximating Pu-238 outputs could be achieved by 1980.

Thulium-171

This isotope is not available today. But there are two ways of producing it: (1) irradiate thulium (Tm-169) at a very high flux (2×10^{15}), and (2) irradiate isotopically pure Er-170.

Present sources of thulium and erbium are small. However, thulium is as prevalent in the earth's crust as bismuth, and only slightly less so than mercury and iodine. Furthermore, it is more prevalent than cadmium and silver. Based on contacts with an existing vendor, it is estimated that with a firm requirement and with about a year of lead time, between three and four tons of thulium could be provided. Similarly, erbium is not particularly rare in nature, for it is over twelve times as prevalent as thulium and is only slightly less common than boron. Other common elements less prevalent than erbium include tantalum, bromine, antimony, thallium, mercury, iodine, and bismuth.

However, as in the case of neodymium, drastic efforts would be necessary to achieve significant production increases of thulium. Again, the most likely source would be the wet phosphoric-acid industry, from which about 25 tons per year of thulium might be obtained. In addition, at least 250 tons of erbium might be recovered, which might yield about 25 tons of **Er-170** as the purified isotope. Irradiating 25 tons of **Er-170** could probably yield about 25 kg of **Tm-171**, or about 5 kW. High flux exposure of thulium (25 tons) might yield 15 kg of **Tm-171**, or 3 kW per year. Again, as in the case of **Pm-147**, thulium production could almost double every year, so that the **thermal** output from this isotope could also approximately match that from an "all out" **Pu-238** production program in 1980.

Thallium-204

Although this isotope is available today, it cannot be made in concentrations suitable for use as a heat source.

Thallium is a byproduct from the metals processing industry and is available at about 15 tons per year. The supply, though presently limited, could very likely be greatly increased. Although, the amount of this increase can only be guessed at, an increase by a factor of 10 via "all out" efforts throughout the mineral industry would not seem unreasonable, especially since there is only one producer in the U.S.

Thallium is more prevalent in the earth's crust than mercury, iodine, bismuth, cadmium, or silver. Irradiation of natural thallium and separation of relatively pure **Tl-204** by a yet-to-be-discovered process should yield about 5 kg of **Tl-204** per ton of **Tl-203** in natural thallium. Natural thallium is about 33 percent **Tl-203**. Thus about 25 kg of **Tl-204**, which could generate 17 kW, might be produced annually from existing sources. By delivery of virgin thallium and recycle of the old, this production level could be approximately doubled every year. By 1980, such development could more than match the thermal output from anticipated **Pu-238** sources.

Thulium-170

Another heat-source isotope which should be mentioned is **Tm-170**. This isotope has properties which would seemingly make it a direct substitute for **Po-210**. Its physical, chemical, metallurgical, compatibility, and biological properties indicate

it to be far superior to Po-210. The only disadvantage is that Tm-170 requires about 150 percent more shielding than Po-210. Where a one kilowatt Po-210 source requires one inch of lead from shielding, the same size source with Tm-170 would require 2.5 inches. However, for large and some small sources, the problem of shielding may not be significant. Furthermore, Tm-170 should be significantly cheaper to produce than Po-210 at any level of production. A production reactor can, per unit of available space, produce about 30 times more radioisotopic thermal energy from Th-169 than from bismuth. In addition, this could be done on a large scale without disrupting the existing market; in contrast, a large bismuth irradiation program could have an undesirable impact on existing bismuth economics since a large fraction of the existing supply which normally serves other common and important uses would be consumed in any large-scale polonium program. The impressive significance of thulium as a heat source may be seen from the fact that it can be obtained without any chemical processing, and that thulium irradiated in a Hanford reactor could readily yield a significant amount of energy at discharge. The anticipated demand for Po-210 is expected to vary from 140 kW in 1968 to a high of 1820 kW in 1972. A small number of tons of thulium could supply these requirements.

Other Isotopes

A large number of other isotopes winnowed from the isotope charts which have a potential application were examined in addition to those discussed above. These are summarized in Table II.

Table II

CHARACTERISTICS OF RADIOISOTOPIC HEAT SOURCES

	Co ⁶⁰	Sr ⁹⁰	Ru ¹⁰⁶	Cs ¹³⁷	Ce ¹⁴⁴	Pm ¹⁴⁷	Tm ¹⁷⁰	Tm ¹⁷¹	Tl ²⁰⁴	Po ²¹⁰	Th ²²⁸	U ²³²	Pu ²³⁸	Am ²⁴¹	Cm ²⁴²	Cm ²⁴⁴
1. Watts/Gram (Pure) ^(a)	11.4	0.95	33.1	0.42	25.6	0.33	12.1	0.2	0.75	141	170	4.4	0.56	0.11	120	2.65
2. Half-Life, Years	5.3	27.7	1.0	30	0.78	2.6	0.35	1.9	3.8	0.38	1.9	74	86	458	0.45	18
3. Estimated Isotopic Purity, %	10	50	3.3 ^(b)	35	18 ^(c)	95	10 ^(d)	90 ^(d)	20 ^(e)	95	95	85	80	90	90	95
4. Compound Form	Metal	SrTiO ₃	Metal	CsCl	CeO ₂	Pm ₂ O ₃ ^(f)	Tm ₂ O ₃	Tm ₂ O ₃	Tl ₂ O ₃	Metal	ThO ₂	UO ₂	PuO ₂	Metal	Cm ₂ O ₃	Cm ₂ O ₃
5. Active Isotope in Compound, %	10	24	3.3	28	15	82	8.8	79	17.7	95	83	75	70	90	82	86
6. Watts/Gram Compound	1.7	0.23	1.1	0.12	3.8	0.27	1.07	0.16	0.13	134	141	3.3	0.39	0.1	98	2.27
7. Density of Compound, g/cm ³ actual or 90% TD	8.7	3.7	12.2	3.6	6.6	6.6	8.5	8.5	9.0	9.3	9	10	10	11.7	9	9
8. Power Density, W/cm ³ Compound	15.2	0.94	13.4	0.42	25.3	1.8 ^(f)	9.1	1.36	1.20	1210	1270	33	3.9 ^(g)	1.17	882	20.4
9. Volume for 2 kW of Heat, cm ³	132	2130	149	4760	79	1120	220	1470	1650	1.65	1.58	61	513	1710	2.27	88
10. Availability Annual kW _t , 1968 ^(h)	Avail.	Avail.	Poten.	Avail.	Avail.	Avail.	Avail.	Poss.	Poss.	Avail.	Poten.	Poten.	Avail. ⁽ⁱ⁾	Avail. ⁽ⁱ⁾	Poten.	Avail. ⁽ⁱ⁾
11. Type of Radiation (Major)	γβ	βx	γβx	βyx	βyx	β	β	β	β	a	αγ	αγ	a	a	an	an
12. Shielding Required ^(j) [MeV significant β or γ]	Heavy (9.5) [1.33 γ]	Heavy (6) [2.26 β]	Heavy (9) [3.35 β]	Heavy (4.6) [1.17 β 0.67 γ]	Heavy (10.2) [2.88 β 2.18 γ]	Minor (1) [0.23 β]	Moderate (2.5) [0.97 β]	Minor (0.3) [0.1 β]	Minor (1) [0.77 β]	Minor (1) [0.8 γ]	Heavy (11.2) [2.6 γ]	Heavy (11) [2.6 γ]	Minor (0.1) [0.04 γ]	Minor (0.7) [0.06 γ]	Minor (0.4) [0.04 γ]	Moderate (2) [0.04 γ]
13. Biological Hazard, MPC, μCi/cm ³	3 x 10 ⁻⁹	10 ⁻¹⁰	2 x 10 ⁻⁹	5 x 10 ⁻⁹	2 x 10 ⁻⁹	2 x 10 ⁻⁸	10 ⁻⁸	4 x 10 ⁻⁸	9 x 10 ⁻⁹	7 x 10 ⁻¹¹	2 x 10 ⁻¹²	9 x 10 ⁻¹²	7 x 10 ⁻¹³	2 x 10 ⁻¹²	4 x 10 ⁻¹¹	3 x 10 ⁻¹²
14. Estimated Future Price \$/g (Pure) [Present Price]	280 [452]	20 ^(k) [28.40]	120 ^(k)	10 ^(k) [10.9]	50 ^(k) [477]	75 ^(k) [186]	121	100	67	2800 ^(l)	6600	1540	300 ^(k)	100 ^(m)	2000 ^(m)	170 ^(k)
15. Estimated Future Price, \$/W	16	21	5	24	2	220	10	500	100	20	40	350	540	910	17	64
16. Curies/Gram (Pure)	1130	142	3394	87	3180	928	6048	1150	458	4500	4100 ⁽ⁿ⁾	114 ⁽ⁿ⁾	17	3.25	3320	81
17. Curies/Watt	65	148	102	207	126	2788	500	5750	610	32	24	26	30	30	28	29
18. Minimum Cost, \$/kWh _e ^(o) [for 1 year mission]	15 (5)	6.2 (10)	23 (1)	6.6 (10)	11 (1)	350 (3)	122 (0.4)	1150 (2)	114 (4)	235 (0.4)	89 (2)	95 (10)	138 (10)	208 (10)	180 (0.4)	22 (10)
19. Spontaneous Fission: Half-Life, Years	---	---	---	---	---	---	---	---	---	---	---	8 x 10 ¹³	4.9 x 10 ¹⁰	1.4 x 10 ¹³	7.2 x 10 ⁶	1.4 x 10 ⁷
20. Availability in 1980, kW _t ⁽ⁱ⁾	MWs	850	6500	850	10,000	40	---	---	---	---	---	---	>400 ^(p,q)	118 kg ⁽ⁱ⁾ 174 ^(q)	---	29 ⁽ⁱ⁾ 110 ^(q)

(a) Includes contributions from daughters at equilibrium (thermal watts).

(b) From power reactor fuels, 20,000 MWd/ton, 1 year after discharge.

(c) Isotopic purity is not expected to exceed 3% at time of recovery from power reactor fuels.

(d) Tm¹⁷⁰ at this concentration is expected on Tm¹⁶⁹ irradiation without need for isotopic separation.(e) Tm¹⁷¹ requires production from isotopically separated Er¹⁷⁰ or irradiation of Tm¹⁶⁹ at very high flux.(f) Tl²⁰⁴ at this concentration is probable only with isotopic separation or irradiation at very high flux.(g) Metal may be preferred; m.p. = 1080 °C. [F. Weigel, Angew. Chem. 75 451 (1963) and expected power density = 2.3 W/cm³]

(h) Does not include the usual 1:1 void volume allowance for helium pressurization with this and all other securely encapsulated alpha emitters.

(i) From proposed plant at Hanford (see HW-77770).

(j) From spent fuels from civilian power reactors (95,000 MW_e in 1980).

(k) Except for shielding against neutrons. Number in parentheses indicates approximate inches of lead shielding required for 1 kW source for 10 mR/hr at 1 m. (See ORNL-3576 except for promethium and americium).

(l) From large civilian power reactor spent fuel processing and isotope recovery facility, after 1970 (see BNWL-25).

(m) Upward revision of minimum costs shown in "Holsted-Albaugh Hanford Diversification Report," 1/15/64, p. 13.

(n) Anticipated price with large scale reactor fueling with plutonium (Pu at \$10/g).

(o) Alpha disintegration curies including daughters at equilibrium.

(p) At terminal or lowest specific power (W/g) at 5% thermal-to-electrical conversion efficiency.

(q) For comparison, power from chemical batteries (specifically flashlight batteries) is calculated to cost \$100/kWh_e.(r) From accumulated Np²³⁷ from spent fuels from civilian power reactors (920 kg neptunium recoverable in 1980).

(s) With recycle of plutonium and americium.

(t) Users of this chart should note the date of issue. Re-issues are made at least two times each year to include most current data.

a Alpha
β Beta
γ Gamma
n Neutron
x Penetrating bremsstrahlung

Extension of Table VI, page 52, HW-76323, REV1,
"Radioisotopic Heat Sources," 10/15/63,
C. A. Rohrmann,
Battelle-Northwest
Richland, Washington

3/1/67^(r)

Flux Traps For Isotope Production

In the case of the flux traps, three basic cases were considered: (1) a D_2O trap for applications requiring high thermal flux levels at ambient neutron temperatures, (2) a light water trap constructed so as to emphasize resonance absorption provided sufficiently little moderator can be employed and yet provide adequate cooling, and (3) a graphite trap operating at high temperatures to provide a thermal flux whose neutron temperature is as close as possible to low-lying resonances of some transuranium isotopes.

D_2O Traps

Two types of D_2O traps were investigated: D_2O regions with and D_2O regions without fuel in the regions. To achieve a significantly high flux in the **non-**fueled case, a special driver region appears necessary outside the D_2O trap. This region would act as a buffer and epithermal-neutron-buildup region above and beyond that achieved by the regular lattice since the original reactor lattice would likely constitute the majority of the reactor load.

Somewhat more complicated is a D_2O trap that is driven in the D_2O region. To take full advantage of the high flux capabilities afforded through D_2O , a D_2O coolant is necessary. This requires recirculating the coolant, which appears difficult but which is not out of the question, as will be described.

After considerable discussion with DUN engineers, it was decided that the simplest system to consider would be a horizontal cavity involving the removal of graphite from the appropriate **number** of lattice positions, but with some alteration of the penetrations in the rear face.

In the case of the undriven D_2O trap, the traps need not be large, **i.e.** no greater than 2 feet in diameter. However, to achieve higher fluxes, a special driver loading surrounding the trap is necessary. The trapped flux could be maximized by increasing the power density in the driver region. But to increase this power density would require increasing the enrichment and possible shortening the length of the driver charge. A more effective driver region could be created by using localized overboring to remove graphite from the driver region.

In the case of the power-driven D_2O trap, a horizontal tank with several stub holes, corresponding to the existing reactor lattice spacing, passing through the rear openings would be inserted from the front. Then regular process tubes could be placed through this region and D_2O could be used in the tank. This provides considerable moderation with D_2O since the lattice would be that associated with the graphite reactor (7-1/2 to 8-1/2 inches), and several target holes could be inserted between the tubes for such low heat-generating targets as bismuth. If once-through cooling with light water were used in this system, the flux achievable would be limited and the possibility of leaking water into the D_2O , with a resulting increase in reactivity, would have to be considered. Thus, one comes back to the simplicity of non-heat generating D_2O target traps, the flux of which is built up, with additional driving being achieved in the water-cooled tubes in the regular graphite lattice whose buffer zone of overboiled tubes immediately surrounds the trap. As indicated previously, a D_2O cooled, power-driven D_2O trap is not ruled out completely, because it could be obtained, without compromising front-face accessibility, by using a re-entrant system from the rear face. The system would, of course, require that the recirculating D_2O system heat be exchanged into H_2O or air. An adequate D_2O handling system for many contingencies would be required, including both a holdup system for the roughly filtered and moderately contaminated D_2O and a provision for disposal of the filtrate.

Light Water Trap

A light water trap was specifically considered for achieving hard spectra at fluxes above those that could be attained simply by filtering out the "thermals". These hard spectra would be gained by using a tight-packed, water-lattice, power-generating region with voided region for targets if the fuel itself did not contain the target material. Visualize an extra large process tube, perhaps 8 to 10 inches in diameter passing through the reactor. In this region, for a 3- to 6-foot length, would be placed a UO_2 tight-packed lattice containing the target regions. With such a system, a high flux of fairly hard spectrum and fluxes considerably higher than those of light water moderated power reactors could be achieved. However, to adopt such a system for a Hanford reactor may create a serious charge-discharge complication. Nonetheless, this system has the virtue of employing water cooling with once-through operation, front-face accessibility, and so forth. Several versions of these systems were examined, as were some

unpowered, light-water traps. An unpowered, light-water trap has the disadvantage of the flux dropping off. For this reason, it does not appear to be a profitable option.

Variable-Spectrum, Light-Water Trap

Since there will be a large number of light-water power reactors, a light-water, power-driven trap in a production reactor must, in order to be unique, supply needs that power reactors cannot supply. It appears that water-moderated and water-cooled regions in a Hanford reactor can achieve flux levels that are a factor of 10 greater than those employed today in light-water reactors. It is of equal, if not of greater, importance that the front-face accessibility of a Hanford production reactor permits the simple adjustment of the moderator when a given load is being irradiated. Specifically, the spectrum can be adjusted by inserting tapered metallic wedges from the front face to displace some of the water.

There may be real merit in using such light water trapping for some of the steps required in producing Cm-248 or even Cf-252. However, such light-water trapping was not pursued during the course of this study with as much emphasis as perhaps is now merited in view of the current interest in californium.

Several studies were devoted to spectral shifting caused by water displacement in tight-packed water lattices. In particular, a study was undertaken of plutonium burning, the results of which are appended. It is important to recognize that there is a limit to using a light water coolant and moderator to achieve an ever-hardening spectrum and at the same time to cool the load. As one reduces the amount of water as a moderator, one will eventually lose cooling capacity. However, recognizing that we are not talking about a critical reactor for these regions, but rather a system driven from the rest of the reactor, and a Hanford reactor at that, one can consider fairly short regions of perhaps only two feet long, and thus a large pressure drop could be accommodated over this short region to get very high water flow rates. Hence, for economic operation, a much tighter lattice can be employed than that of a conventional light-water power reactor whose fuel lengths are 9 to 12 feet or so to provide economic operation. Because of the large pressure drop capability of the existing Hanford pumping system, the tight lattice could produce surprisingly hard spectra and at the same time quite high power densities.

It is, of course, recognized that a light water lattice is limited in its ability to harden the spectrum and cool the system simultaneously. If greater hardness beyond this is desired, then one can consider D_2O , which provides essentially the same coolant capability as light water, but with significantly less moderation. Using D_2O , of course, would involve a recirculating cooling system, which poses significant problems for Hanford, although it would be possible to preserve the accessibility of the front face by introducing and withdrawing the coolant via the rear face.

Graphite Moderated Traps

After some discussion and computations, it was concluded that the only real incentive for a graphite trap would be to achieve high neutron temperatures. Consequently, considerable study was devoted to the relative reaction rates achievable at high neutron temperatures as compared with hard spectra and/or low neutron temperature thermal spectra. (This study constituted a large amount of the latter activity under MFC-12 and was reported in supplemental reports. The important thing is that at high neutron temperatures the effective reaction rates of many isotopes of interest, specifically U-236, Np-237, Pu-242, and several curiums, appear to be enormously improved. For completeness, a cryogenic irradiation was considered as well as irradiation at temperatures as high as 3500°C. A temperature of 3000°C was considered a limit for solid and liquid materials because adequate densities above this temperature cannot be achieved by simple means. Hopefully, irradiation targets can be successfully prepared for 2000°C; and, eventually, 3000°C. Analysis at these temperatures appeared to be important because of low-lying resonances of some of the transuranium and transplutonium isotopes. If a thermal spectrum (even its upper tail) can reach these resonances, higher reaction rates can be achieved than those of some "resonance systems" that are limited by target self-shielding. At first, the high-neutron-temperature target most specifically considered involved the production of Pu-238 and such production, indeed, holds great promise. However, the potential value of Cf-252 has received most attention and while almost no quantitative data are available, our study results suggest that it may be worthwhile to pursue high-neutron-temperature targets as a means of producing Cf-252, or at least of producing some of the intermediate isotopes leading to Cf-252.

Augmented Fluxes Surrounding the Fast-Test Module

An outgrowth of a fast-flux test module is a high thermal flux in the area surrounding the critical fast reactor module. The estimate appears to be something like 10^{15} nv. Because this will be a relative large volume, its use as an irradiation area should be considered, both at low neutron temperatures and at the elevated neutron temperatures previously discussed. In the case of low neutron temperatures, it is reasonable to consider a D_2O region rather than a graphite. In addition, the flux in a D_2O region may rise to something like 10^{16} , provided the target absorption is not too great.

Achievable Flux Levels

Many computations were made with respect to the design of the various traps to see what sort of flux arrangements might be achieved. It soon became apparent that identifying a most economical configuration for each situation would not be possible in the course of the study. It was very difficult to come up with a simple figure of merit that served to "sort out" the worth of various approaches. A simple figure of merit is the peak flux level in the trap divided by the average flux level in the driver region, or, more specifically, the flux level in the remainder of the reactor. Comparisons were made with this ratio for different types of traps: graphite, D_2O , and water. For each of these types, ratios were compared for various trap sizes and for various moderator densities in the driver. Comparisons were also made with the density of driving material in the trap, namely, U-235.

The resulting ratios tabled below (Tables III, IV, and V) show pretty much that for any given moderator in the trap at a given density, a maximum flux in the trap will occur as a function of trap size. (This maximum flux depends principally on the "neutron age" and parasitic absorption of the trap). Or, for a given trap size, the maximum flux will occur at a certain density. Generally speaking, the maximum flux ratio was achieved for a given trap size with the lower moderator densities in the driver region. These results are not unexpected. However, one does achieve some combinations that may be of interest. For example, a ratio of 8 to 10 for the flux in the trap over the flux in the driver is achieved at such low graphite densities in the driver as 0.2 gram per cubic

TABLE III
FIGURES OF MERIT TO COMPARE

1. ϕ_T/ϕ_D vs. Type (Gr, D₂O, H₂O).
2. ϕ_T/ϕ_D vs. Trap Size (0.5 ft., 2.0 ft., 6.0 ft.).
3. ϕ_T/ϕ_D vs. Mod. Density of Driver.
4. ϕ_T/ϕ_D vs. Density of Trap.
5. ϕ_T/ϕ_D vs. U-235 in Trap.

TABLE IV
EFFECT OF TRAP SIZE AND DRIVER MODERATOR DENSITY ON ϕ_T/ϕ_D

		<u>Graphite Density In Driver (g/cm³)</u>			
<u>Size</u>		<u>1.638</u>	<u>0.819</u>	<u>0.409</u>	<u>0.205</u>
D ₂ O	0.5	2.71	3.66	4.29	4.50
	2.0	2.81	4.72	6.67	8.00
	6.0	1.96	3.53	5.45	6.95
GR	0.5	1.49	1.82	1.93	1.89
	2.0	1.41	2.28	3.12	3.65
	6.0	0.85	1.47	2.28	2.96
H ₂ O	0.5	0.31	0.70	1.55	2.91
	2.0	0.07	0.18	0.39	0.75
	6.0	0.03	0.06	0.13	0.25

TABLE V
EFFECT OF TRAP DENSITY ON ϕ_T/ϕ_D *

		Trap Size		
<u>Trap Density</u>		<u>0.5</u>	<u>2.0</u>	<u>6.0</u>
D ₂ O	1.0	3.66	4.72	3.53
	0.5	3.02	4.32	3.80
Gr	1.638	1.82	2.28	1.47
	0.819	1.65	2.19	1.94
H ₂ O	1.0	0.70	0.18	0.06
	0.5	1.54	0.42	0.15

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* Density of graphite in driver is 0.819 g/cm³.

centimeter, as compared with 1.6 for ordinary graphite. Trap density, of course, is not as important since it is a function of the diameter of the trap. A certain amount of U-235 in the trap for purposes of building up the flux is perhaps warranted if very large fluxes are desired. Relatively slight amounts of U-235 in the traps can build fluxes up by factors of 10 or 20 over the drivers. However, the fact remains that this would also involve the cooling problems previously discussed and would have to be examined in great detail. Also, some consideration was given to removing essentially all of the moderator in a portion of the driver surrounding the trap region (i.e. leaving some regular process tubes in place without graphite surrounding them).

Reaction Rates

The criteria of flux ratios may be too limited because the real consideration boils down to the reaction rates that can be achieved. This then brought us back to a more thorough comparison of the trade-offs between flux levels, spectral hardness, and neutron temperature as applied to each reaction of interest. (If no resonance absorption exists for a given isotope, then a hard neutron flux is of no value.) This further brought us to the fact that there is a gross lack of information on magnitude and location of resonances for isotopes heavier than Pu-242. If it is desired to produce Cf-252 in a truly optimum fashion, then considerably more data are required. Or, as a minimum, one would desire to begin some well planned experiments to irradiate Pu-242 for various trap concepts and then examine the yield of Cf-252 and its precursors as a function of exposure time. (This may be an easier and more relevant approach than attempting to measure the cross sections as a function of energy for each isotope involved).

The relative reaction rates for various neutron temperatures and spectral hardnesses are reported in BNWL-CC-2000-AOPG. This represents several thousand computations, and a selected sample is contained in Table VI. These data, by themselves, are extremely useful to indicate the direction of research. However, the trade-offs associated with selecting an optimum are not obvious, especially when costs are involved. Many attempts were made to develop a pragmatic system for "optimizing", but far too much judgment and hard computation was involved for that. It was concluded that an optimizing computer program was required to couple with an adequate reactor-physics and cost-code complex so that various

TABLE VI
EFFECT OF U-235 IN TRAP ON ϕ_T/ϕ_D

	U-235/bc	Trap Size		
		0.5 ft.	2.0 ft.	6.0 ft.
D ₂ O	0.453×10^{-4}	9.64	11.30	14.10
	0.543×10^{-5}	10.00	22.50	34.40
	0.543×10^{-6}	3.54	8.95	24.90
Gr.	0.543×10^{-4}	5.01	5.85	7.96
	0.543×10^{-5}	4.00	11.60	21.90
	0.543×10^{-6}	1.71	2.88	6.87
H ₂ O	0.543×10^{-4}	22.20	37.90	52.60
	0.543×10^{-5}	0.52	0.14	0.08
	0.543×10^{-6}	0.321	0.08	0.03

optimal criterion could be pursued. Presumably, the criterion most sought after is minimum cost to produce a given amount of product in a certain time. The system now works well and is described in the document BNWL-991. With the current cross section data, it appears that optimal systems could be computed for: (1) producing from stable targets such isotopes as polonium, thulium-170, thulium-171, and Co-60; (2) producing Pu-238, and (3) producing Cm-242 and Cm-244. As mentioned elsewhere, gross uncertainties in cross section data prevent sufficiently accurate computations for the transplutonium isotopes beyond Cm-244.

Unevaluated Trap

For hard spectrum applications that involve the burning of fissile fuel as, for example, curium production from plutonium, the tightly packed water-lattice flux trap appears optimum. However, for irradiations that do not involve large amounts of heat, and yet require a hard spectrum, other approaches are indicated so as to minimize the costs of altering the reactor preparing driver fuel, etc.

In the case of a low-heat generation system, it is observed that light-water hard spectra are essentially out. Because of parasitic absorption, the flux levels are simply too low, and if a reasonably sized region is required, are too soft. It is obvious that some fissions have to be made close to the region in order to provide a source of high energy neutrons whose energy could then be adjusted by different amounts of moderation. For this application, it might be better to use a minimum or no moderator by removing the graphite from around a group of process tubes, mechanical support being afforded by zirconium brackets. Some of the central tubes of the group could contain the targets. Modest amounts of heat generated in the targets may then be removed by: (1) limited amounts of light water cooling until the spectral hardness is compromised, (2) steam cooling, (3) larger amounts of D_2O cooling than (1) above, and (4) gas cooling or possibly air cooling if contamination is no problem. Adjustments to the neutron spectral hardness could be made by distributing supplemental moderator among the tubes. This approach to a hard neutron spectrum is not conventional because it makes poor use of reactor volume. This is not a problem in a Hanford reactor and this could be an attractive mode of operation. In this case, recirculation of coolant is required for D_2O and possibly for gas cooling. It is considered here because relatively small amounts of heat are involved and because the system could be fed from the rear face and thus preserve accessibility to the front face.

Combination Systems

A system briefly considered is a combination of the foregoing, namely, special test holes designed for different jobs, all in the same reactor. One could use the adjustable resonance absorption regions employing the variable moderator displacement characteristic of light water for some of the steps to californium, for example, burning plutonium so that several resonance absorption regions could be utilized and considerable energy liberated. This step **could** be followed by successive irradiations in regions of either high or low neutron temperature as appropriate. The ideal, perhaps would be to move targets from one irradiation environment to another without rejacketing them or processing them chemically. Whether or not this reconstitution of targets could be low enough in cost remains to be determined. It is particularly relevant that for some of the steps in a chain production sequence, a **Hanford** reactor "as is" may be **ideal**. For example, Am-241 should not be irradiated at too high a flux in order to minimize fission of the intermediate isotope, Am-242. It should also be **recognized** that if holes for these target irradiations were put in, they would also be available for materials testing. Thus it appears that a Hanford reactor could indeed be outfitted to do a wide variety of jobs, many of **which** cannot be done in any existing reactor in the AEC complex or perhaps anywhere else today.

V. SUMMARY OF FLUX TRAPPING

Observation

To summarize: In the area of test reactors, the potentially large test volumes available in the Hanford reactors are unique. The fact that these test volumes can provide fairly flat neutron fluxes over relative long lengths is very definitely unique. Thus one could have long test "slots" available for simplifying certain experiments. Or one could have a fast flux testing region with an extremely high thermal flux surrounding the fast testing region. Or one could have a high neutron target temperature region, a hard spectrum target region, and a very high thermal flux in a D_2O trap. Moreover, it does not appear unreasonable to have all these capabilities available in one reactor, even if not all at the same time. Such a reactor would be highly unique within the AEC complex.

It is suggested, in keeping with the simplification of the basic Hanford reactors, that the following be pursued:

1. Once-through cooling system and no power generation, and thus no contamination of the front face allowing front-face accessibility 100 percent of the time. For the few cases where recirculating coolant is necessary, it is hoped that a re-entry coolant system from the rear face could be used.
2. Horizontal rather than vertical target holes. It is hoped that a minimum reactor containment will be necessary.
3. No further consideration be given to power generation.

Containment, Recirculation and Heat Sinks

On the assumption that MFC-12 would cause a Hanford reactor to be started up from standby, some consideration was given to whether or not MFC-12 revisions should require special effort to (1) prevent radioactivity in the coolant system from contaminating the Columbia River, (2) provide a high degree of containment, (3) reduce thermal loads to the river, and (4) provide for a meltdown in the reactor. But very little work was done in these areas beyond a review of

previous work. It was concluded that no special effort would be made on MFC-12 revisions to change the once-through coolant system or to grossly alter the containment systems. Since some of the possibilities considered are of interest and may be of value in the future, they are set down here as a part of the MFC-12 report.

Recirculated Coolant

Several systems of **recirculating** coolant were reviewed. Most of these had been discussed during other programs dealing with radioactivity of the Hanford environs. There are several recirculation studies in the literature, some by General Electric, and later some by DUN, which generally concluded that the cost of recirculating coolant was prohibitive. During MFC-12, some potentially **lowered-price**, although less elegant and less satisfactory, recirculating systems were discussed and briefly considered. One of these would involve a large holdup in the recirculating system so as to reduce activity in the front face. Hopefully, the heat exchangers involved could be very low-priced **low-heat-transfer-surface** types exchanging to water in essentially open canals at near atmospheric pressure or under pressure barely enough to provide an adequate velocity to increase the heat transfer coefficient in the sink side. It was observed that possibly one of the lowest priced sources of tubing would be aluminum-irrigation pipe at low internal pressures. A sandwich design using epoxy glue and aluminum plates was also studied (a paper patent was discussed). In the recirculating systems considered, it was assumed that once the major portion of the heat was removed from the recirculating coolant, it could be ponded back in the existing clear-well systems or perhaps even into a portion of the main tanks now here initially employed from the river pump houses.

A problem associated **with** coolant recirculation is the gradual buildup of radioactivity in the recirculating system to the extent that shielding is required in the front face and throughout the system. It was observed during work with an old Hanford reactor that a modification to an ordinary **recirculating** process could be considered; namely, that upon shutdown of the reactor, the system could work on a once-through basis for cleanup to reduce the buildup if radioactivity levels were not too high, during power operation, the reactor would be restored to a recirculating coolant.

Modified Once-Through Systems

Other considerations of radioactivity in the coolant were briefly considered. Among these is the backyard lake, with its potential problem of blowing dust that is slightly radioactive due to the drying out along the edges of the lake and the associated wave action which could wet a larger higher elevation than would actually be covered by the lake. Whether or not a riprapping with large rock along the edge of the lake could conquer this problem was not determined. A long canal was also briefly considered and rejected, as was a certain cataphoresis process for reducing the amount of radioactivity water to the river by a factor of 10 or so. Perhaps one of the most relevant suggestions, also rejected, was that with restart of an old reactor, water might be run through the flocculation step after, rather than before, passing through the reactor, in an attempt to remove much of the radioactivity before the water is put back into the river. This would require a modification of the processing system because some flocculation would be required before passing water through the reactor. This is required because raw river water could involve serious filming problems which could enhance radioactivity problems. The observation was also made that in the double reactor areas (a single reactor area could be connected by canal with another area to achieve the same thing), water could be run through the flocculation step before and after passing through the reactor, if both water plants were used.

Initial Conclusions

In any event, it was finally concluded that there are a sufficient number of possibilities that could be employed (if deemed advisable) to modify the once-through systems, and so no special considerations were made in MFC-12 reactor modifications toward a recirculating system, even though the special holes or regions of the reactor worked up for MFC-12 could possibly be used for recirculation more easily than the other regions of the reactor. It was observed again and again throughout the MFC Study that there are many desirable features of a once-through system; in particular, the accessibility of the front face.

Thermal Effects

Along with special considerations of radioactivity there are those of thermal effects. In a truly reasonable sense, the "thermal pollution" problem of the Columbia River due to the Hanford reactors is perhaps more emotional than real.

If demands are sufficient, it does appear that many of the ideas discussed in the past by DUN (and formerly by GE) to alleviate the thermal pollution problem could be employed. In particular, a backyard lake, a long "seeping" return channel, and cooling towers are means of reducing thermal pollution. The reduction of thermal pollution by using a power generating cycle on the old reactors was discussed with consultants, but the fact remains that the amount of energy to be converted into electricity is simply too small to seriously affect the thermal pollution problem.

Thus, again it was decided specifically not to adjust the ideas of MFC-12 in order to provide a recirculation system for radioactivity or thermal pollution, or both, but rather to go on the assumption that the once-through system discharging heat to the Columbia River would be the major means of operating the Hanford reactors, even if modified by a concept from MFC-12. It is recognized, of course, that there may be certain recirculation loops for testing, such as gas-cooling or **sodium-cooling**, which rather obviously need not discharge radioactivity or even heat to the river. Although in the main, as the study progressed, it appeared that neither of these approaches would not constitute a large market for the sorts of energy and neutron capabilities of a Hanford reactor.

Containment

Along with considerations of recirculation from the standpoint of radioactivity and thermal contamination, the overall containment problem was briefly examined again to see if some special considerations of MFC-12 should be undertaken to either simplify or alter containment ideas. There are several aspects of this that come to the fore immediately. One of these is the containment sphere. These appear to all observers to be infeasible from the standpoint of the near impossibility of inserting a sphere section underneath the existing reactor. The next containment aspect usually considered is that of strengthening the existing buildings. This is a stopgap measure and in the case of the old Hanford reactors would be grossly insufficient due to the inherent weaknesses in these buildings. For evacuating off-gas during routine operation, reactors are provided with stacks, and since the reactor locations are relatively remote, a system providing containment to allow scheduled discharges is hardly necessary unless the advance of civilization makes remoteness less and less real.

Another concern for containment is unscheduled steam evacuation or other bursts of gas, both of which imply the use of a containment system such as a sphere. If MFC-12 does not introduce any grossly new nuclear effects on water coolant above the boiling point, it is difficult to see a large gaseous burst resulting from the Hanford reactors. Because of the expense of a system such as a sphere or other high pressured containment, an additional containment capability does not seem reasonable. However, recognizing that MFC-12 was to be directed toward starting up a shutdown reactor, it was felt that some speculation should be given to some alternative methods of containment. Some of these considered included circus tents over the reactor working at reduced pressure, the exhaust being conducted to some other system for containment. Rather than circus tents, astrodomes and geodesic domes appeared to offer more satisfaction. The astrodome type structures were excluded because of cost, and of those polled only the geodesic domes appear low enough in price to serve the function. An order-of-magnitude price of a million dollars was quoted by Mr. Buckminster Fuller over the telephone for a 400-ft diameter dome that could withstand 200-mile per hour winds and 60 pounds per square foot external loading. This system would work under partial pressure, be exhausted by blowers up the stack for routine operation and exhausted into a "bag" for containment holdup. An astrodome, at somewhat significantly larger cost could be acquired for handling larger external pressures, but essentially no net internal pressure can be considered unless a closed system such as a tank is used as a dome "takes off". In common with all of these are problems of sealing around the system, no total solutions were identified but it was believed that a sufficiently large blower system and a sufficiently sealed system could be made. Several thoughts were given to handling the off-gas for temporary holdup. Among these were (1) the inflation of plastic bags lying in trenches (Canadian idea), (2) passing the gases over ice to condense steam and lower gas pressures (Westinghouse suggestion), and (3) passing gas through a water-well system (General Electric suggestion). Another method was that of providing a very large containment volume working at reduced pressure for a containment system that would be activated only in the case of an event. The idea was to search for the lowest priced piping or other associated low-pressure vessel that can be secured (for example, culvert pipe made galvanized ribbed steel for use underneath roads) and use it for containment. One idea was to lay a large number of these culvert pipes at a slight incline in the

immediate vicinity of the reactor and under normal operations to keep these pipes filled with water. The culvert pipes would be connected through a large header system to the reactor containment system. The containment system normally works at reduced pressure, with blowers discharging through the stack. In case of an event, the blowers system to the stack would be closed off and the water in the culvert pipes would be released at its lowest point, so that it could then serve as the exhaust function from the containment sphere. The idea of such an approach is to provide a very high-capacity, partial-pressure system at a relatively low price.

Containment Unnecessary

In going through all of these systems, when one checks the amount of gas and so forth that would have to be handled, the fact still remains that if the reactor operation is maintained such that the majority of the water coolant is **below** the boiling point of water, then no large (pv) energy release would occur in the event of a pipe rupture or other untoward event. Thus even the need for containment system becomes difficult to justify, although a modest investment could possibly provide a spirit of adherence far beyond that currently achieved. This, of course, assumes that the once-through coolant system to the river could be temporarily checked during the event so that gross amounts of radioactivity would not be **con-**veyed to the river. This then gave additional emphasis for MFC-12 considerations to not pursue a power generating **cycle**, even one **using** a refrigerant working fluid much less a high-pressure steam system.

Meltdown

Meltdown was another consideration, especially because of the possibility associated with MFC-12 for introducing unusual fuel loads in the reactor. It has been pointed out that with an uncontained reactor system using horizontal tubes, one of the real dangers **is** not that of a loss of coolant and evolution of steam and gases but rather a meltdown within the reactor **slowly** releasing so much radioactivity throughout the environs that no containment system could handle it. Or, possibly, meltdown with material running out of the reactor tubes **onto** the front or rear face and reconstituting a critical mass. The thinking extended to considerations that possibly the lowest priced approach might simply be to build

ramparts around the reactor building (essentially a dike) and in an emergency to flood the reactor. If a geodesic dome were put on top of the rampart the sealing problem associated with a system like this would possibly be enormous. So enormous that it may be most difficult to ever work the system at a truly reduced pressure because the leaks inward would be so great that no visible reduced pressure would exist, although the exit of gases from the system up through the stack would be very great. This would be a significant task for the blower system although there would be a large dilution in the gases going up the stack simply because of the large amounts. However, with the availability of vinyls and other plastics, it is very possible that significant sealing could be achieved.

Conclusions Regarding MFC-12

It was concluded that in behalf of MFC-12 a once-through cooling system would be adhered to, that no energy would be generated into electricity, that the coolant temperature would be purposely limited to as low a temperature as possible, and that preferably the bulk of the reactor coolant would be below the boiling point of water. It was also concluded that front-face accessibility would be maintained, and that the possibility of more extensive containment would merely be acknowledged, with no special steps being taken in the MFC-12 concepts to simplify these steps above and beyond that historically necessary for standard operation of Hanford reactors.

APPENDIX ARAMPARTS AND NOVEL CONTAINMENT

As AEC production reactors are diverted from weapons applications, it is natural that expenditure rates to provide engineered safeguards may be more difficult to maintain. Thus, the physical plant may require augmentation to provide more intrinsic **containment**.

Hanford reactor operation for nondefense missions may well be interpreted as requiring containment systems in the spirit of the safeguards interpretations applied to power reactors. The pressure vessel approach appears too expensive to install about an existing facility with the possible exception of N Reactor. For maximum future flexibility, it is important to stimulate creative thinking on ways of resolving this potential safeguards problem. A study of unusual methods by DUN and PNL personnel was proposed. The following are examples only of the sort of thing to be examined by such a study.

Cursory attention has been given to containment and provision of cooling in case of potential meltdown. Since considerable land is available, it may be possible to provide effective containment by an appropriate combination of steps. For example:

1. The alluvial material characteristic of the site can be used to provide ramparts around the reactor building to provide capability of last ditch flooding of the reactor to prevent fission product release from complete meltdown. This, in turn, can cut down on the requirements of the containment system just as the sprayed-water basin systems to be applied to commercial boiling water reactor does.

2. The overall containment might be a geodesic dome (approximately \$1,000,000 for a 400-ft. diameter unit similar to the DEW line units which handle 60 lb/ft² snow load and 200 miles per hour winds) standing upon the top of the ramparts. This unit would be maintained at negative pressure routinely with gases normally going up the stack.

3. Inflatable bags could be laid out in the desert for emergency storage of radiation gases above limits for routine stack discharge (these could be laid in trenches dug for the rampart construction).

4. Plastic may be used to seal the ramparts and dome joints in view of the need to provide a negative pressure.

5. The characteristics of water discharge to the river must also be dealt with. This may require at the extreme, complete recirculation of reactor coolant water. During the initial considerations for this proposal, it has been observed that a fairly low priced recirculation system may be possible if the reactor coolant temperature is maintained below 100°C. Thus, rectangular water channels made with folded metal may be used rather than a tube heat exchanger.

6. On the other hand, more extended water holdup and/or more selected additions to the water may be sufficient. It is anticipated that prior Hanford studies and the USAEC, Division of Production suggestions in this regard could be re-examined as part of this proposal. These include backyard lakes, extended canals parallel to the river, or outright channeling of the river.

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