

The Aircraft Reactor Experiment-Physics¹

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The fluoride of a fissionable material dissolved in molten fluorides of other cations can serve as the fuel of a circulating-fuel nuclear reactor. These fluorides have a slowing-down power about one-half or one-fourth of the slowing-down power of dense graphite. The resonance escape probability depends strongly on the cation but is always less than that of carbon. The consequences of these properties for various reactor applications are discussed.

Techniques for critical experiments for molten fluoride reactors have been developed, and the physics aspects of operation of the ARE have been analyzed. Operation of the ARE demonstrated that molten-fluoride reactors have strong negative temperature coefficients, mainly as a result of fuel expansion. The ARE was shown to be very stable and to be a slave to the power load. No ¹³⁵Xe poisoning was found in the ARE, and the radioactivity of the fuel after removal from the reactor was less than it would have been if all fission fragments had been retained. The loss of delayed neutrons by fuel circulation modified the in-hour equation but not the stability of the ARE.

GENERAL REMARKS ON MOLTEN FLUORIDE REACTORS

As pointed out in a preceding paper by R.C. Briant and A.M. Weinberg (1) almost all proposed molten salt reactors use fluorides as the salts. Among the favorable properties of fluorine is its low absorption cross section, which at thermal energies is about the same as that of beryllium. Further, its average logarithmic energy loss per collision, 0.10 as compared with 0.16 for carbon, is the highest obtainable in an anion of acceptable chemical properties. It was also pointed out by Briant and Weinberg that this average logarithmic energy loss per collision is not so high as the reactor physicist might wish and that reactors moderated entirely by fluorine would have very high critical masses.

A contributing cause of the high critical mass is to be found in the cations with which the fluorine is combined. Beryllium with its low absorption cross section and great slowing-down power is highly desirable. However, the high viscosity of molten beryllium fluoride limits the permissible concentration of this compound in the salts to about 30 mole per cent.

It is regarded as practical to separate lithium isotopes, and ⁷LiF is another desirable constituent of the fuel solvent salt. However, the thermal absorption cross section of ⁷Li, 33 mb, is not really very small; it is ten times that of carbon. The scattering cross section, 1.4 b, is exceptionally small, and therefore the microscopic slowing-down power, $\xi\sigma$, of ⁷Li is no greater than that of fluorine. Furthermore, the ⁶Li content of the lithium would have to be reduced from its natural value of 7.5% to 0.0035% before the contribution of the ⁶Li to the lithium absorption cross section would be reduced to the level of the contribution of the ⁷Li. Such a reduction of the ⁶Li concentration can hardly be attained by isotope-separation methods, but the ⁶Li would gradually burn out if the salt were used over and over in the reactor.

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TABLE I
PROPERTIES OF FLUORIDES AND CONVENTIONAL MODERATORS

Composition (mole %)	^6Li atoms per ^7Li atom	Resonance escape probability to 0.075 eV	Slowing-down power (cm^{-1})
D ₂ O		0.9997	0.18
Graphite		0.995	0.07
69 LiF-31 BeF ₂	0	0.95	
	0.0001	0.89	0.038
	0.001	0.46	
50 NaF-50 ZrF ₄		0.58	0.021
46.5 LiF – 11.5NaF – 42 KF	0	0.14	
	0.0001	0.13	0.018
	0.001	0.059	

Table I shows, for comparison, the estimated resonance escape probabilities and macroscopic slowing-down powers of D₂O (density, 1.1), graphite (density, 1.85), and various fluoride mixtures. The resonance escape probability refers to the case of an infinite space filled with the material in question; in other words, the moderation is supplied only by the material itself, not by additional moderators.

From the column on the resonance escape probability it may be noted that even the idealized lithium-beryllium fluoride salt, containing no ^6Li , has a lower resonance escape probability than that of D₂O or even that of graphite. The salt loses, in epithermal absorption, about 5% of the fission neutrons. Hence, a fluoride-salt homogeneous reactor cannot be used as a thermal breeder, no matter how large it is. However, a lithium-beryllium fluoride fuel, with a small quantity of ^6Li atoms per ^7Li atom, would achieve criticality in a homogeneous thermal reactor and could be used as a burner or converter. A lithium-beryllium fluoride fuel with more than 0.001 ^6Li atoms per ^7Li atom would not achieve criticality in a homogeneous thermal reactor, and neither would sodium-zirconium fluoride or the alkali-fluoride fuel mixtures.

The slowing-down powers of the fluorides listed do not vary much—only by a factor of 2. This small variation is due to the above-mentioned fact that the cations, even beryllium and lithium, make only a limited contribution to the neutron moderation; and the fluorine, which contributes, in any case, a large fraction to the slowing-down power, is represented by about 3.6×10^{22} atoms/cm³ in the fluoride salts, with no more than 25% variation, up or down. The slowing-down power of the fluorides is less than that of graphite and, of course, very much smaller than that of D₂O. Any homogeneous, bare reactor would be very large, but fluoride salt without fissionable material could be used as a reflector to reduce the size of the core.

Another means of reducing the size of the core would be the use of high concentrations of fissionable material, which would cause the fissions to occur predominantly at higher energies and thus give the neutrons less chance to leak out. The high concentrations also allow the fissionable material to compete effectively with the absorbers in the salt so that fluorides other than the ^7Li -beryllium fluoride could be used or a somewhat higher ^6Li content could be tolerated. An embodiment of these ideas was described at the end of the paper by Briant and Weinberg.

Briant and Weinberg also mentioned that the somewhat mediocre properties of the fluoride salts as moderators suggested the use of some of the conventional moderators for the slowing down of the neutrons, the salts being then largely confined to the role of carrier of the fissionable material. In other words, heterogeneous reactors offer some promise. In such reactors, relatively small quantities of the salt are used, and therefore the parasitic absorptions in the salt are, within

limits, unimportant, and salts other than ${}^7\text{Li}$ -beryllium fluoride can be employed. In view of the high temperatures required by the melting points of fluoride fuels, aqueous moderators appear to be impractical, and therefore beryllium metal, beryllium oxide, and graphite can be considered. Beryllium and beryllium oxide cannot be used in contact with molten fluorides, and when used as moderators in fluoride systems they must be protected.

All the structural alloys that can withstand the molten salts and which can be used for the protection of the moderator contain large amounts of nickel (which has a sizeable thermal absorption cross section of 4.6 b) and some other metals with large absorption cross sections. The thickness of the protective layer must, of course, exceed a minimum amount because of corrosive attack. Also, the volume-to-surface area ratio, or the linear dimensions, of the moderator blocks cannot be large because of self-shielding and similar effects and because of thermal stresses resulting from neutron and gamma-ray heating of the moderator blocks. Hence, the amount of absorbing alloy needed to protect the moderator cannot be reduced very much by increasing the sizes of the blocks. This problem is not serious in burners or low-conversion-ratio converters, but, for the extreme neutron economy demanded by breeders, the protective alloys and, hence, beryllium and beryllium oxide are not likely to succeed.

Graphite is believed, but not yet proved, to be compatible with the fluorides. The calculation, mentioned by Weinberg and Briant, regarding a ${}^{235}\text{U}$ -Th system with a breeding ratio of about one refers to a reactor moderated by uncanned graphite.

AIRCRAFT REACTOR EXPERIMENT

For a molten fluoride reactor experiment, such as the ARE, it was clearly desirable to use a moderator to supplement the moderation by the fuel salt so that the size and uranium investment of the reactor would not be too large. Beryllium oxide was chosen as a moderator. The only fluoride fuel available at that time was a mixture of sodium, zirconium, and uranium fluorides in which the uranium was enriched to 93.4% ${}^{235}\text{U}$. The beryllium oxide moderator blocks were protected from the fuel by the Inconel tubes in which the fuel flowed. As stated in the preceding paper by Bettis et al. (2), the shape of the BeO blocks was fixed by the availability of hexagonal blocks obtained for a solid-fuel reactor.

The usual reactor statics investigations of core size, reflector thickness, temperature coefficients, etc., included multi-group calculations for which IBM machines and a predecessor of the Medusa and Eyewash codes were used. Since these methods are now well known (3), they will not be described here.

CRITICAL EXPERIMENTS

In order to check the multi-group calculations, a critical experiment was performed with the assembly shown in Figure 1. The array of hexagonal BeO blocks with longitudinal holes in their centers is clearly visible. The axial holes in the central portion, or core, are occupied by Inconel tubes, which were filled with fluoride fuel in powder form to simulate the circulating fuel. The reflector is formed by the blocks with small axial holes for the coolant. One of the instrument holes is also visible in the foreground.

In the critical assembly, the measurements shown in Figure 2 were obtained. These data are presented in lieu of measurements on the ARE, because no means was provided in the ARE to determine the neutron spectrum or the spatial distribution of the neutron flux. The critical assembly differed in some minor points from the actual reactor and was operated at room tempera-

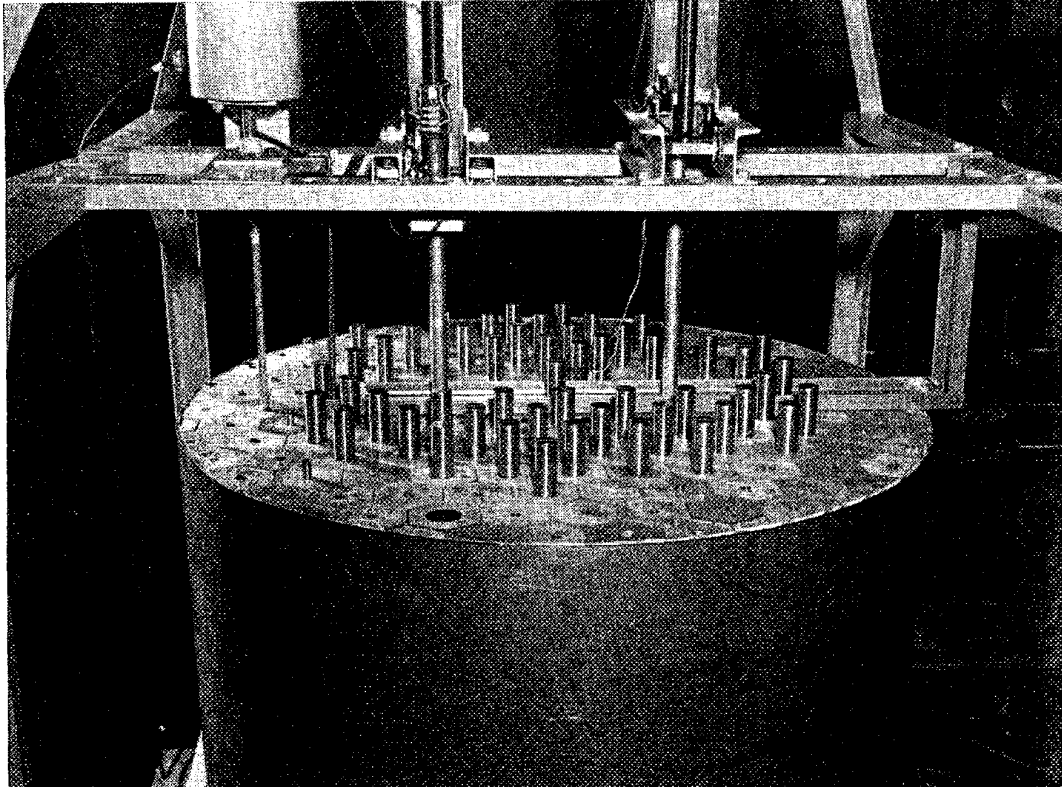


FIGURE 1: Critical Assembly of ARE

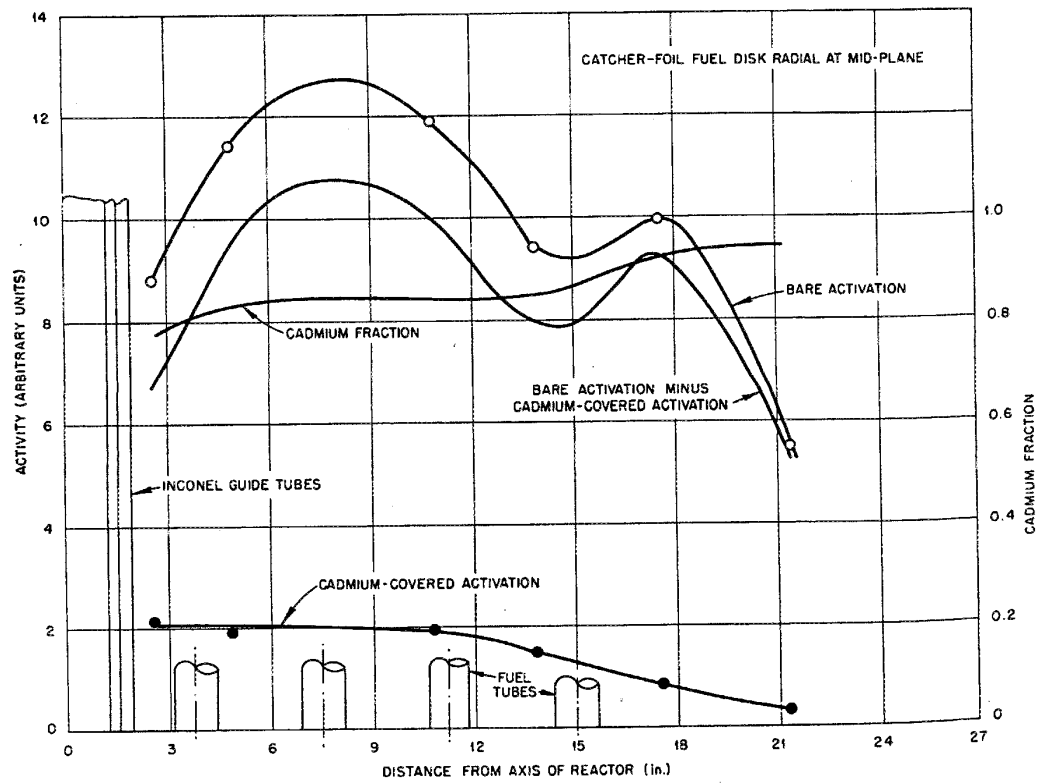


FIGURE 2: Power Distribution in ARE Critical Assembly

ture, but the general features of the measurements are undoubtedly applicable to the reactor experiment. The figure shows the activation of aluminum catcher foils which covered a 1.4375-in-diam (36.5 mm), 0.01-in.-thick (0.25 mm) enriched-uranium disk. The fast flux, as indicated by the "cadmium-covered activation," is highest at the center and falls off rather smoothly with increasing radius; the thermal flux, as indicated by the "bare activation minus cadmium-covered activation," is depressed in the center by the regulating rod mechanism and falls off toward the core reflector interface, but it reaches a peak in the reflector. From the figure it would appear that only about 20% of the activation was due to the fast flux, but the measurements were made with a small foil which did not depress the thermal flux very much. In the fuel tubes of the reactor the thermal flux was considerably reduced and hence a sizeable percentage of fissions was caused by epithermal neutrons. Of course the usual control rod calibrations, measurements of importance functions of various poisons, etc., were performed in the critical experiment.

In the early room-temperature critical experiments, including the critical experiment discussed above, the fluorides, which are solid at room temperature, were used as powders. This made it somewhat difficult to change the uranium concentration to accommodate different loadings. In later experiments, uranium foils and Teflon sheets were used as mockups for the fuel, and it was somewhat easier to change the ratio of uranium to Teflon and, hence, the uranium concentration.

In the uranium foil-Teflon sheet assemblies it is impractical to make the foils less than 0.004 in. thick (0.1 mm). Even for this small thickness, self shielding of the foils is not negligible. This was investigated rather carefully (3) for the case in which the assembly is infinite in extent. Whether this investigation is exactly valid for the case in which the lattice borders on the moderator has to be investigated separately. Since this self-shielding in the lattice is not present in the actual reactor, any error in the investigation would affect the calculation of the critical mass of the actual reactor if the results of the critical experiments are used for the calculation.

More important than the self-shielding of the foil in the lattice is the already mentioned self-shielding of the fuel channels. This self-shielding will always be present in a heterogeneous reactor. In order to minimize the amount of neutron-absorbing alloy around the channels, the volume-to-surface area ratio of these channels, that is, their linear dimension, will be increased until the self-shielding is really serious. A room-temperature critical experiment does not really represent the conditions at high temperature because the absorption cross sections and hence the self-shielding are smaller at elevated temperatures.

The fact that self-shielding is not adequately represented by room-temperature critical experiments is the most important of several reasons why high-temperature critical experiments are desirable. In these experiments, the molten fluoride fuel is lifted by gas pressure into a replica of the planned reactor core, no pumps and fuel circulation being necessary because no heat is generated in the critical experiment. The Oak Ridge National Laboratory is now prepared to perform such high-temperature critical experiments. In these experiments it is also relatively simple to change the uranium concentration by adding a concentrated solution of uranium fluoride in another salt.

In fact, the ease with which the uranium concentration can be changed in a fused salt reactor is an important advantage, since fuel burnup and fission product poisoning necessitate fuel additions during operation of any reactor. Besides, the critical masses of reactors are not easy to predict and a wrong prediction is less serious when it can be compensated for by simple additions of liquid fissionable material than when it requires refabrication of solid elements.

Actually, the critical mass of the ARE was predicted fairly accurately on the basis of the results of the critical experiment. The critical mass of the ARE was 14.9 kg of ^{235}U , which was 0.38 g of ^{235}U per cubic centimeter of fuel. There is a somewhat diffuse upper limit for the desirable

uranium concentration. In sodium-zirconium fluoride carrier this limit is about 0.45 g of uranium per cubic centimeter of fluoride mixture. If this upper limit is exceeded, the melting point and viscosity of the fuel solution gradually become excessive.

TEMPERATURE COEFFICIENTS

One of the most desirable features of the molten salt reactors is the strong negative temperature coefficient. This negative temperature coefficient results in extraordinary stability of the reactor. The most important contribution to the negative temperature coefficient comes from the expansion of the fuel. The fuel salts have thermal volume expansion coefficients of $3 \times 10^{-4}/^{\circ}\text{C}$ (within a factor of 2) that are considerably larger than the volume expansion coefficients of the metals which serve as the walls of the container. Upon heating, the molten salts expand out of the reactor core and leave less fissionable material, and less scattering material, in the reacting zone. If a fraction $\Delta m/m$ of the fissionable material is expelled from the reactor, the reactivity is decreased by a fraction $\Delta k/k$, the ratio $(\Delta k/k)/(\Delta m/m)$ being one-third or less for most reactors. For the ARE the ratio was 0.24. Hence, we might expect on the basis of expulsion of ^{235}U , a negative temperature coefficient of approximately $10^{-4} (\Delta k/k)/^{\circ}\text{C}$ for molten fluoride reactors. In the ARE the negative fuel temperature coefficient actually was greater, $1.75 \times 10^{-4} (\Delta k/k)/^{\circ}\text{C}$, and indicated that effects other than expulsion of ^{235}U contributed to the coefficient.

The "fuel temperature" coefficient discussed above is observed if the reactor power changes fast enough so that only the fuel and not the rest of the reactor changes in temperature. A slow temperature change in which most parts of the reactor reach thermal equilibrium with each other yielded a negative "reactor temperature" coefficient of $1.75 \times 10^{-4} (\Delta k/k)/^{\circ}\text{C}$, which is only about two-thirds of the fuel temperature coefficient.

An attempt was also made to study the influence of the reflector coolant (sodium) temperature on reactivity. The sodium was cooled rapidly to prevent the moderator from following its temperature. In this way, the sodium temperature coefficient was also found to be negative. In another experiment the sodium was cooled so slowly that the moderator also dropped in temperature while the fuel temperature was kept constant. This experiment gave the sodium temperature coefficient plus the moderator temperature coefficient. Since the sodium temperature coefficient was already known, the moderator temperature coefficient could be determined. The moderator temperature coefficient turned out to be positive, and thus the reason for the above-mentioned reactor temperature coefficient being smaller than the fuel temperature coefficient became evident.

Among the various temperature coefficients, the temperature coefficient of the fuel is the most important for the operation of the reactor, since the temperatures of the other components of the reactor do not change rapidly. As mentioned, the negative fuel temperature coefficient resulted in stability of the reactor. Furthermore, the power generated by the reactor was essentially determined by the power extracted. When the power demand was increased, the reactor cooled down a little, the reactivity increased, and the power increased; in other words, the reactor was a slave to the power load. It is quite possible that a reactor of this kind could be operated without control rods.

The effect of the power load on the ARE was demonstrated in a number of experiments, one of which is illustrated in Figure 3. The abscissa, which is to be read from right to left, is the time, in minutes, and the ordinate is the printout from recorders that gave the reactor inlet and outlet temperatures. Initially, the reactor was operated at low power. Then a slow increase in the heat extraction from the fuel was first indicated as a drop in the temperature of the fuel which reached

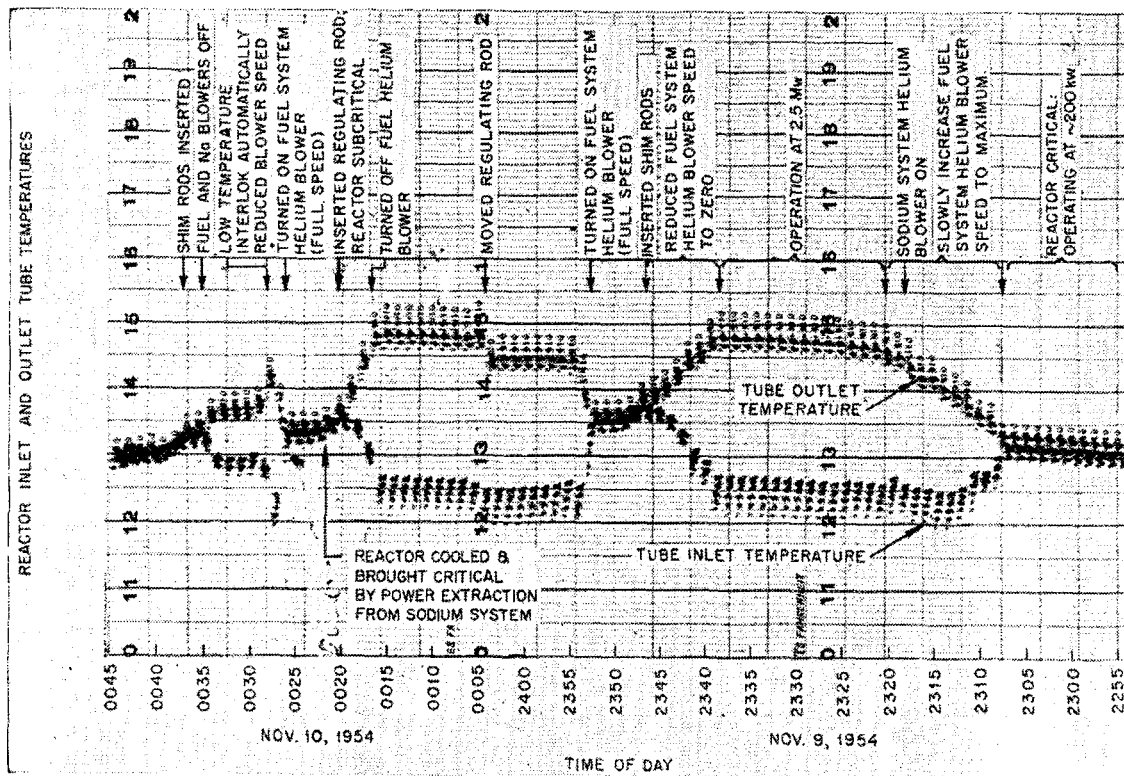


FIGURE 3: ARE inlet and outlet temperatures.

the reactor inlet from the heat exchanger. This change in the temperature of the fuel increased the reactivity and the reactor power, as indicated by the temperature rise at the reactor outlet. The spread of inlet and outlet temperatures corresponds to a power of 2.5 MW. When the heat extraction was again reduced the inlet temperature rose and the outlet temperature fell until the two temperatures became nearly coincident. The control rods did not determine the power output; they only influenced the average temperature. The shim rod insertion dropped the temperature. A rapid increase in the power demand on the fuel system again spread the inlet and outlet temperatures apart, and the full insertion and full withdrawal of the regulating rod depressed and then raised both temperatures simultaneously. Next, the power extraction was stopped and the regulating rod was inserted to make the reactor subcritical.

A demonstration that the reactor could be brought to criticality by the power demand without the use of the rods was then made. Power extraction from the sodium system cooled the reactor to make it critical and then the power extraction from the fuel again caused the spread of inlet and outlet temperatures. Most remarkable was the stability of the power level when the reactor was left alone. The power level showed none of the ripple which it had been anticipated might result from possible mechanical vibration, etc.

ELIMINATION OF ^{135}Xe AND OTHER FISSION FRAGMENTS

The stability made it unexpectedly possible to demonstrate that less than 5% of the ^{135}Xe was retained in the reactor. It had been computed that the xenon poisoning after 25 hr of operation at full power should be 2×10^{-3} in $\Delta k/k$ if all the xenon formed stayed in the fuel until it decayed. This poisoning was less than would have been expected from the usual equations, partly because the fuel spent only one-fourth of the time in the core and was thus, effectively, only subjected to a

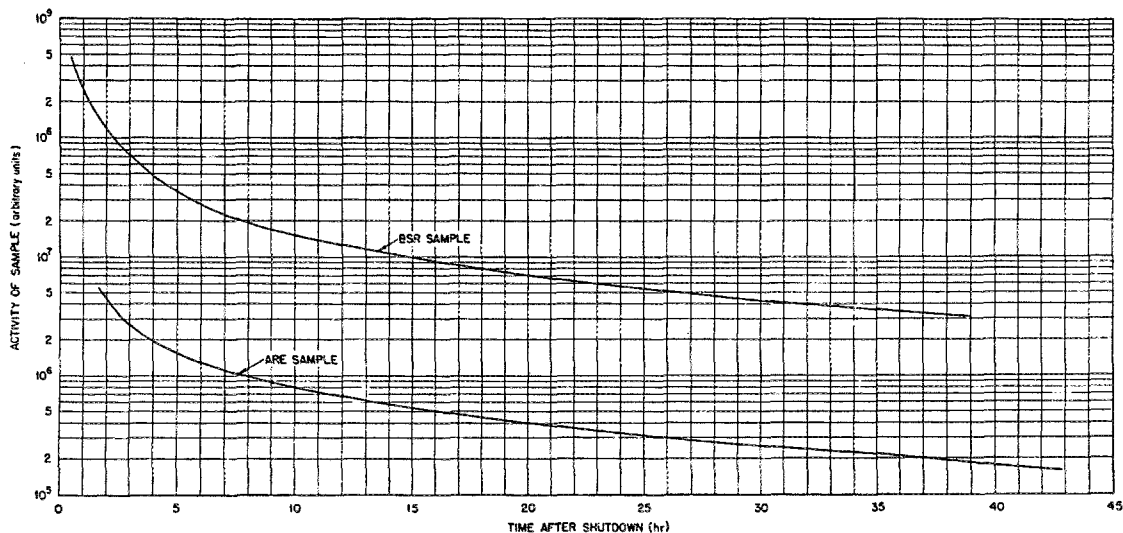


FIGURE 4: Activity of fluoride samples.

quarter of the flux, and partly because many of the neutrons had energies above the large ^{135}Xe absorption resonance. As little as 5% of the computed poisoning would have been detectable, but none was found (4). Hence, in a molten-fluoride reactor there will be very little, or no, xenon poisoning or danger of a positive temperature coefficient due to xenon. Little or no excess reactivity has to be provided to override xenon after a shutdown, at least if the fuel can be circulated before startup.

Obviously, if ^{135}Xe is not retained in the molten salt, other noble gas fission fragments will escape. It is also possible that additional fission fragments will be volatilized from the fuel or will be removed from the reactor by other processes. A considerable amount of the fission fragments disappeared from the ARE fuel, as indicated by the following experiment. A sample of the fuel was withdrawn from the ARE after a shutdown following reactor operation at 27 W for 1 hour. This was early in the experiment, and the reactor had only been operating at very low power prior to this 1-hr run. A similar sample was irradiated in the Bulk Shielding reactor at ORNL under a power history which duplicated the power history of the ARE prior to the withdrawal of the first named sample, except for a proportionality factor of 0.37. The decay of the gamma activity of both samples is plotted in Figure 4.

If the necessary corrections are taken into account, but if it is assumed that both samples retained all their fission fragments, the activity of the Bulk Shielding Facility sample should have been 12.4 times as high as that of the sample from the ARE. Actually, the ratio was 27 at 1 hr and 40 min after shutdown and 17.5 at approximately 38.5 hr. The discrepancy is easily explained by the fact that the Bulk Shielding Facility sample was solid and did not lose any fission fragments, whereas the sample from the ARE was stripped of some of its fission products. Since the most volatile fission fragments include strong gamma-ray emitters with relatively short half-lives (such as 2.77-hr ^{88}Kr and its Rb daughter), it can be explained in a qualitative way why the molten sample curve is depressed relatively more at short times after shutdown.

From the safety viewpoint, the loss of fission-fragment gases is both a blessing and a curse. Since the volatile fragments are given off during operation and can be released, under controlled conditions, or stored where they would not be liberated during an accident, an accident under such conditions could reach essentially only the nonvolatile fragments and could not release much airborne activity. Unless extremely high power densities were used, the fuel would solidify after

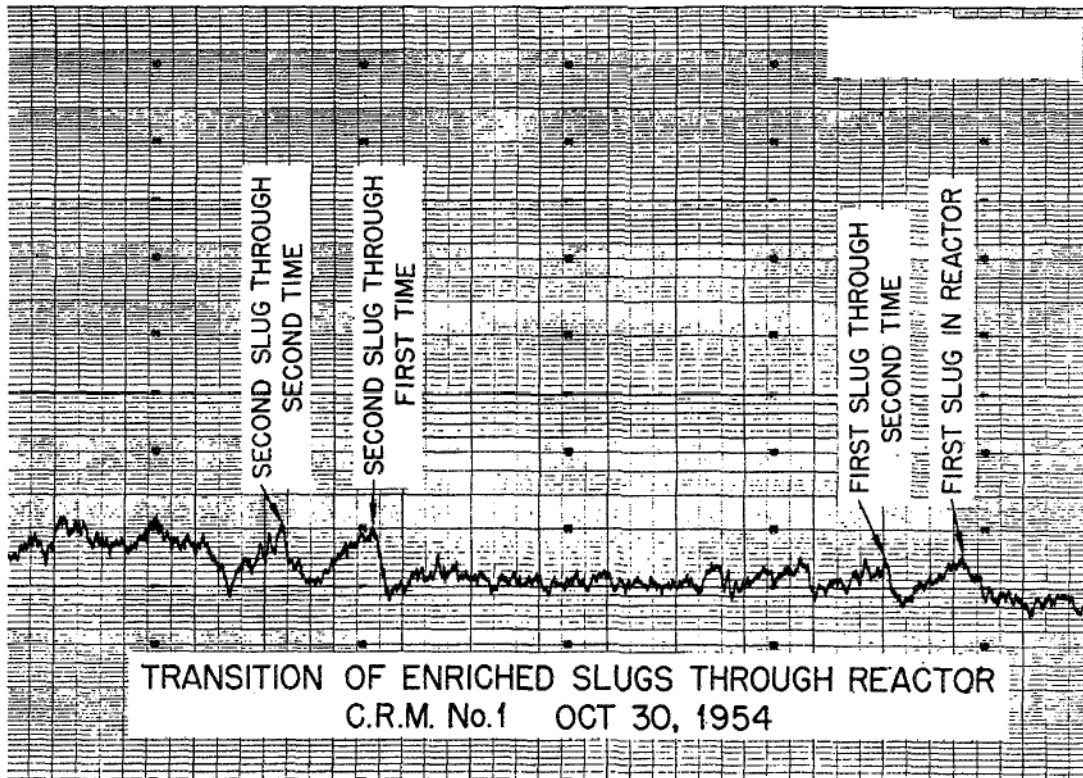


FIGURE 5: Transition of enriched slugs through the reactor.

the accident and occlude any gases that formed. On the other hand, during normal operation the requirements for leak tightness of the system are rather stringent.

CIRCULATING SLUGS

The elimination of gaseous fission fragments was undoubtedly due to a large amount of stirring of the fuel, but, in spite of this stirring, liquid slugs of fuel retained their identity for several passages through the reactor and external circuit. This was first noticed during the approach to criticality, when ^{235}U was added in slugs to barren salt. When the slug of high uranium concentration passed through the reactor the first time, the counting rate of the meter temporarily increased, as can be seen in Figure 5, and it increased again when the slug went through the reactor the second time. A similar phenomenon is shown in Figure 6, where the fuel circulation had been stopped while the reactor was at some low power, and where the pump was started up after a scram. The slug which had been in the reactor during power operation contained delayed-neutron emitters and emitters of hard gamma rays, which can create neutrons by the Be-photoneutron reaction, and every time this slug went through the reactor again, the meter showed a "pip." At least five such passages, could be recognized before mixing and decay of activity washed out the picture. Six or more pips following a scram after a rapid rise in reactor power can be seen in Figure 7. The slug which filled the reactor at the moment of the scram is the most active one, since the power had reached its crest at that moment. The pips offered a convenient way of measuring the time it took the fuel to go through the reactor and external loop, and pumps and flow meters could be calibrated by use of this phenomenon.

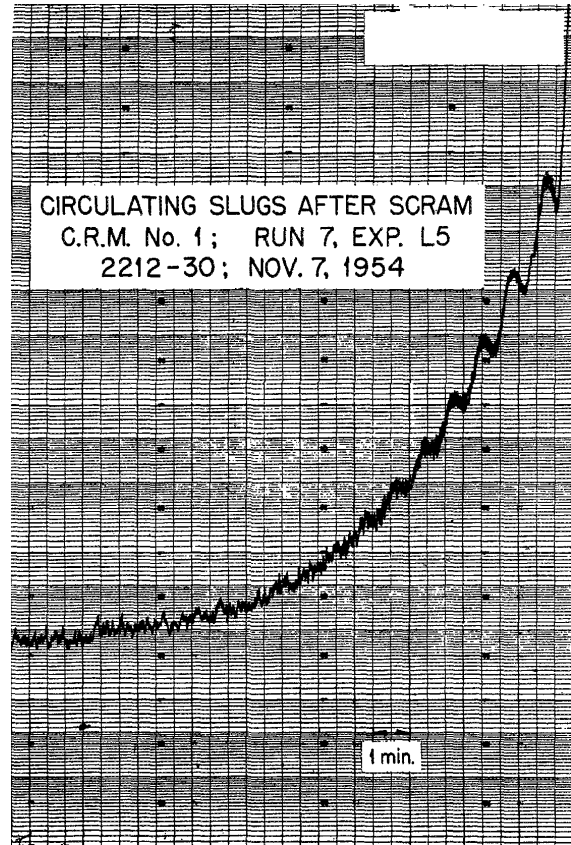
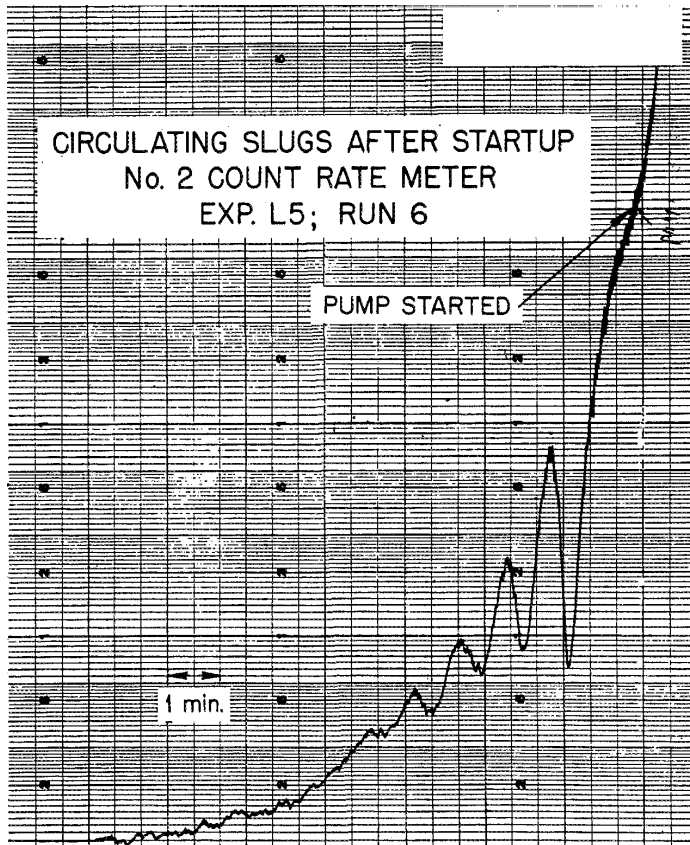


FIGURE 6: Circulating slugs after startup.
 FIGURE 7: Circulating slugs after scram.

LOSS OF DELAYED NEUTRONS

The fact that slugs retained their identity through several passages was gratifying because the inhour equation had been computed under this assumption in advance of the experiment. The inhour equation gives the relation between the reactor period and the excess reactivity, and it depends, obviously, on the delayed neutrons given off in the reactor. In a circulating-fuel reactor some delayed neutrons are lost, because they are born in the heat exchangers, pumps, etc. If the flow rate is very small, these losses are negligible, because the delayed-neutron precursors have time to decay in the reacting zone before being swept out.

The inhour equation is essentially the same as for the stationary fuel reactor. If the flow rate is very high, the birthplaces of the delayed neutrons are evenly distributed over the reactor and the outside fuel circuit, and the effective delayed neutron yields are reduced as compared with the natural yield by the ratio of the fuel volume in the reactor to the total fuel volume. Thus the delayed neutrons are less effective in slowing down the exponential increase of the reactor power, and for a given excess reactivity the circulating-fuel reactor will go on a shorter period than will the stationary fuel reactor. If the flow rate is increased from very low values to very high values, the period corresponding to a given excess reactivity will decrease monotonically.

In the ARE the time spent by the fuel inside the reactor and the total transit time through the complete fuel circuit were intermediate between the mean life of the long-lived delayed neutrons and the mean life of the short-lived ones. The consequence is shown in Figure 8. The long-lived delayed neutrons were largely lost from the reactor and greatly reduced in their effectiveness. A

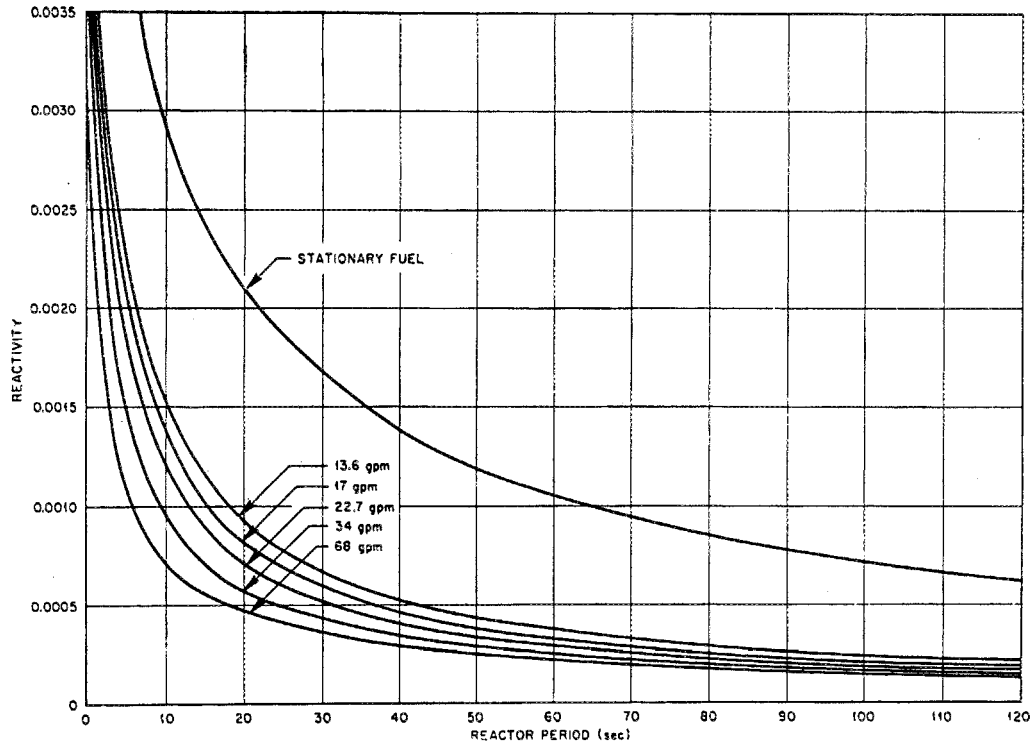


FIGURE 8: Reactivity as a function of reactor period for several fuel-flow rates (reactivity calculated from inhour equation).

relatively small excess reactivity enabled the reactor to increase in power without waiting for the long-delayed neutrons. The small excess reactivity gives a surprisingly short period. On the other hand, the delayed-neutron precursors with short mean lives have time to decay in the reactor, and short-lived delayed neutrons are quite effective. A relatively large excess reactivity has to be introduced to override them and to bring the reactor on a very short period. This behavior is shown in Figure 8 for different flow rates.

The fact that small excess reactivities are sufficient to give short periods caused some misgivings prior to operation of the experiment, but the experiment dispelled these fears, largely because of the negative temperature coefficient.

At some time prior to the experiment it was feared that the loss of the delayed neutrons might result in a loss in damping of any power oscillations of the reactor. However, it was possible to demonstrate mathematically (5) that the circulation of the fuel tends to damp oscillations, and the operation of the experiment did not show any tendency toward oscillations.

REFERENCES

1. R.C. BRIANT AND A.M. WEINBERG, *Nuclear Science and Engineering* **2**, 797 (1957).
2. E.S. BETTIS, R.W. SCHROEDER, G.A. CRISTY, H.W. SAVAGE, B.G. AFFEL, AND L.F. HEMPHILL, *Nuclear Science and Engineering* **2**, 804 (1957).
3. J. H. ALEXANDER AND N.D. GIVEN, ORNL-1925 (1955).
4. J. BENGSTON, ORNL-CF 57-5-19 (1957).
5. E.S. BETTIS, W.B. COTTRELL, E.R. MANN, J.L. MEEM, AND G.D. WHITMAN, *Nuclear Science and Engineering* **2**, 841 (1957).
6. W.K. ERGEN, *Journal of Applied Physics* **25**, 702 (1954).