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CHEMICAL, PHYSICAL AND REACTIVITY CHANGES
IN A FULL REACTOR E-N MELTDOWN

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CHEMICAL, PHYSICAL AND REACTIVITY CHANGES
IN A FULL REACTOR E-N MELTDOWN

R. Nilson

SUMMARY

The events from a chemical standpoint following a total loss of coolant disaster will not be altered in the melting reactor by the introduction of N metal. The inter-diffusion of uranium and aluminum will be the dominating reaction, causing the blockage and tying up of the lithium in UAl_3 which does not melt until after the uranium does. Pressure from the swelling UAl_3 will extrude uranium-aluminum and lithium into graphite weep holes and block interfaces.

The migration of lithium by vaporization will not become appreciable until well over $2000^{\circ}C$, well beyond the time when uranium and UAl_3 have melted. The eventual result will be a diffuse distribution of uranium, lithium, and aluminum in the lattice.

The E-N pile has a larger excess over required control capacity than the uranium provided the large reactivity poison tied up in the lithium is not lost. Compared to the natural uranium pile, the gain of reactivity on loss of coolant is less and the net temperature coefficient in the dry pile remains negative to higher exposures. Furthermore, permanent pile poisoning during meltdown is accomplished via two mechanisms both lithium and uranium redistribution in the lattice produce large negative reactivity effects.

INTRODUCTION

Maintenance of adequate coolant flow to the Hanford reactors is basic to the nuclear safety of the plant. Though the sudden and complete loss of coolant to a reactor is considered highly improbable, the occurrence cannot be ruled out as an impossible one, no matter how reliable the backup systems or how stringent the operating standards are. Acceptance of this fact dictates an operating philosophy and procedure which will not increase the magnitude of the inevitable damage in the event of the maximum credible disaster - total loss of coolant.

The operation of the eight production reactors therefore is governed by two very important safety criteria, namely the speed of control and total control criteria. The E-N loading in terms of these criteria is actually further within the limits imposed by the control criteria than the normal plutonium production piles. The E-N hazards report² adequately discusses the details of the comparison.

However, in the E-N loading a potential source of a very large reactivity gain exists which is not present in the normal uranium loading. The fact that approximately 15 per cent of the dry reactor's reactivity is tied up in the low-melting metal, lithium, naturally gives concern that a supercritical state can occur in the melting reactor if the lithium leaves the pile. Our earlier conclusion² that the removal of lithium from the melting pile is highly improbable has been considered in finer detail³ and it is the purpose of this report to discuss the chemical and physical processes which are expected to occur in the melting pile and what the effects of these changes are on the reactor's reactivity.

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DISCUSSION OF THE CHEMICAL AND PHYSICAL CHANGES³

In an a priori analysis of what might happen in an E-N load following loss of coolant, we are indeed fortunate in a backhanded sort of way to have experienced the K West incident in tube 4669 (HW-35821)² and (HW-35822)⁴. This was essentially a full scale experiment such as one would like to perform in order to answer the questions which might be asked. In addition, the supporting laboratory experiments conducted to establish just what had corroborate the analysis.

Perhaps the best lesson learned here is that less emphasis must be placed on the water and air reactions with the very reactive aluminum, and uranium, at least under the conditions of tube 4669. These appear to be of very minor consequence compared to the damage wreaked by the diffusion of uranium into the aluminum to form UAl_3 . This compound forms in a spongy, voluminous manner and the resultant swelling can completely block the coolant annulus. It is only fair to point out, however, that tube 4669 did not have free access of air. In a worst credible accident such as discussed here, it is altogether possible that free access of air by diffusion and/or convection might be possible. Under these conditions much more aluminum oxide, uranium oxide, and lithium oxide could be formed. These are all high-melting solids and would not distill or migrate to any appreciable extent. The net effect of extensive oxidation would be to retain the light metals in place by the formation of oxides. The worst condition would obtain in an inert atmosphere or in vacuum, for under these conditions most metals will melt and flow readily.

In the case of water loss and heat build-up within the reactor, two phenomena occur simultaneously both radially and axially from the uranium. The first is heat transfer. Fission product decay energy causes an increase in temperature. In the center of the reactor the process is almost adiabatic. The source of heat is the uranium. The radial sinks are Al-Si bond material, 2-S aluminum can stock, gas annulus, 72-S process tube, and graphite stock, in that order. The expected results are melting of the various alloys and subsequent flowing. However, the second phenomena occurring is diffusion of the uranium. This becomes very rapid above 550° C, in the neighborhood of the Al-Si melting point. The result is the formation of UAl_3 , a high melting compound. Apparently, diffusion of uranium and subsequent formation of UAl_3 can occur at a faster rate than the temperature can rise, with the result that the uranium can actually melt and flow before any aluminum alloys can melt and flow. The aluminum alloys may thus be firmly fixed in their original positions in the reactor assembly by the migration of uranium. Even if the alloy should, in part, melt and flow from its position within the process tube, it is difficult to see how it could travel far from its initial position without encountering uranium with which it would react to form the intermetallic compound.

After the uranium melts, the pressure created by the expanding UAl_3 can extrude the molten uranium out of any hole or weak place in the structure and between the graphite blocks and into the graphite weep holes as so ably demonstrated by the photographs in HW-35821. Unless coolant is restored prior to UAl_3 formation, no coolant will be able to be restored. If coolant can be restored, the primary hazard which exists in putting the water back in, is the flash steam hazard. Accelerated corrosion of hot aluminum will occur, but the water is cold and will cool the metal rapidly before this corrosion would be sufficient to cause real trouble. In the event that bare LiAl has somehow been exposed, the action of cold water will be to generate some hydrogen along with that already being generated by the aluminum corrosion. A review of the literature,^{5,6} on metal-water reactions makes an explosion hazard seem unlikely when water is passed over the hot metal. This is in contrast to the situation where liquid metal is added to water.

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DISCUSSION OF THE CHEMICAL AND PHYSICAL CHANGES (CONT'D)

In the event, that a channel or many channels have been blocked by UAl_3 , we shall be very interested in trying to predict the course of events. The situation at temperatures up to around $1350-1400^\circ C$ have been observed and probably quite accurately tell the whole story. The addition of one four-inch $LiAl$ piece to every seven eight-inch uranium pieces, will make just that much more aluminum to react with the uranium and tie it in place a little longer.

In the process of forming UAl_3 it is highly probable that the lithium will be either physically trapped or possibly chemically bound within the matrix. There is also the possibility that some of the lithium will not be so fixed but will remain as an alloy whose melting point will decrease as the lithium concentration increases. In this event, flow of lithium might take place, but with the blockage of flow passages by expanding UAl_3 and molten uranium it is difficult to imagine much gross movement occurring. The lithium is 3.5 weight percent of the aluminum. Its maximum vapor pressure can be estimated by Raoult's law which states:

Vapor pressure of lithium in the molten alloy = the mol fraction of lithium in the alloy x the vapor pressure of pure lithium at the same temperature.

The mol fraction of lithium in the piece is roughly 0.1. The vapor pressure of pure lithium reaches one atmosphere at about $1400^\circ C$. Therefore, at $1400^\circ C$ the vapor pressure of lithium will be 0.1 atmosphere or less. This vapor will diffuse only slowly through the surrounding gas at atmospheric pressure. It will immediately dissolve in any aluminum which does not have an equilibrium value of lithium already. The migration should be quite slow. As it permeates the graphite pores it will absorb on the graphite, further slowing the migration. At an ill-defined upper temperature in the neighborhood of $2000^\circ C$ it will react with graphite to form a carbide which, however, has a short temperature interval of stability and decomposes again within a few tens or hundreds of degrees according to the work of Tucker and Moody.⁷ Around $2000^\circ C$ aluminum and $LiAl$ will begin to boil and will be distilled to the cooler fringes of the reactor assembly through any pressure relief holes or cracks. Meanwhile, however, the UAl_3 will have melted and will flow to cooler regions in the gravity field. The flowing mass will probably be a rather homogeneous uranium, aluminum, silicon, lithium melt at these temperatures.

The tritium, hydrogen, and helium contained in the N pieces will begin to evolve from the N metal at the time that this metal reaches about $500^\circ C$. These slugs will also start to swell at this time and might add to the blockage of the channel which has already occurred because of UAl_3 . The gases will have essentially no chemical effect under these conditions and will probably just act to keep the atmosphere somewhat more inert. The tritium and hydrogen will react chemically with any bare uranium at temperatures below $400^\circ C$ but it is doubtful whether any of this type of uranium will be around. Except for the explosion possibility if this gas gets mixed with air, no new hazard is introduced. These gases will seep through the reactor rather rapidly and will escape to the reactor gas system.

All of the molten metals flowing in contact with graphite will react to form more or less stable carbides, but probably only on the surface. It is doubtful whether such a layer would move more than one-fourth-inch into the graphite perpendicular to the surface in a week of exposure.

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DISCUSSION OF THE CHEMICAL AND PHYSICAL CHANGES (CONT'D)

It is probably very safe to conclude that once UAl_3 melts, the melt will flow as a rather homogeneous melt of uranium, aluminum, silicon, and lithium, with some changes of composition occurring as movement and time progress.

Following the melting of UAl_3 , UAl_2 , and other alloys, the resultant behavior will be strictly a function of the mechanical damage to the reactor of interest. Little would be learned of most laboratory type experiments. The KW incident demonstrates that uranium and aluminum will both flow especially under the pressures exerted by the expanding UAl_3 . The pores of the graphite can also be filled with Al or $LiAl$. If any material comes out of the reactor first, it will probably be uranium.

One question which may require an experimental answer remains. This is the following: when the hot uranium begins to diffuse axially, moving first through the $AlSi$ bond layer, then the one-fourth-inch thick end cap, then across the interface between the E and the N metal, through the thick end cap of the N piece and finally into the N metal itself, will it selectively extract aluminum from the lithium-aluminum alloy, and cause the formation of low melting, high lithium alloy? If this happened, it might be possible to cause lithium alloy to melt and flow. Even under these conditions it would be difficult to see where it might flow since all channels will probably be blocked and filled with uranium or UAl_3 .

DISCUSSION OF THE REACTIVITY CHANGES

Before describing the reactivity effects corresponding to the physical and changes just discussed in the preceding section, it is worthwhile to review the loading pattern and the physics parameters for the E-N loading.

The proposed loading pattern is discussed in the E-N hazards report²; briefly the pattern is a fringe enriched-fringe poisoned type which is an improvement over a normal uniform loading. The central zone of the reactor contains a striped load of 4 inch N slugs ($Li-Al$) and 6.5 inch enriched uranium slugs (0.9%) in a ratio of 1 to 6. Surrounding this core is ring of enrichment, which in turn is surrounded by a ring of poison, in this case $LiAl$. It is sufficient for these analyses to restrict ourselves to just the central zone of the reactor; therefore, the physics parameters are for a normal E-N loading, namely, a striped load in the entire reactor.

The lattice parameters, temperature coefficients, water loss reactivity changes and equilibrium xenon poisoning are given in Table I for both a natural uranium solid fuel element loaded pile and the proposed I & E E-N load. These data are for a BDFH type reactor. It is noted that the multiplication factor for the E-N loading is less than that for the natural loaded pile. This is partly a consequence of the lower reactivity of the internally and externally (I & E) type slug. This loss in k_{00} can be brought back with either sufficient fringe enrichment, or a slight adjustment in the E-N slug ratio, depending on which is most economically attractive.

The constants, f , p , η , ϵ and k_{00} are for the cold, clean reactor. Note the low thermal utilization attesting to the large poisoning effect of the lithium. The resonance escape probability, p for U^{238} is also higher in the E-N lattice, due to the reduced U^{238} nuclear density. Thus the Pu^{239} conversion is lower than in the normal uranium loading. However, the tritium, expressed as gram-equivalents of

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DISCUSSION OF THE REACTIVITY CHANGES (CONT'D)

plutonium, makes the total initial conversion ratio (ICR) considerably larger than in the natural load. A portion of the conversion ratio gain, however, is due to the higher resonance absorptive properties of the I & E slug over that for the solid slug.

The metal temperature coefficient is smaller in the E-N load, due again to less U^{238} in the lattice. The graphite coefficient is also smaller in the I & E E-N pile because of 1) a smaller neutron blocking effect in the I & E due to reduced coolant annulus thickness, 2) a decreased effect of parasitic absorption in the graphite due to increased U^{235} concentration and the nearly black target N slugs, 3) a smaller temperature variation in η due to increased U^{235} content.

TABLE I PHYSICS PARAMETERS (BDFH)

	<u>Nat. U Solid Load</u>	<u>E-N I & E Load (.94%)</u>
f	.866	.769
p	.879	.891
ϵ	1.039	1.034
η	1.327	1.450
$k_{\infty} = \eta \epsilon pf$	1.049	1.039
ICR	.974 gm/MWD	1.039 gm/MWD
Metal coefficient	-1.2 ih/°C	- 1.0 ih/°C
Graphite coefficient ($\bar{E} = 0$)	+ .23 ih/MW	+ .17 ih/MW
Δk , water loss (cold, clean)	+ 750 ih	+ 680 ih
Δk , water loss (hot, $\bar{E} = 500$)	+ 250 ih	+ 160 ih
Equilibrium xenon	975 ih	1000 ih

In a dry, clean reactor the graphite temperature coefficient is negative. However, as plutonium builds into the fuel, the coefficient eventually turns positive. For the E-N reactor, higher U^{235} concentration delays the onset of the inversion in sign of the graphite coefficient due to the larger U^{235} competition for neutrons in the enriched uranium.

The change in reactivity on loss of coolant in the E-N reactor is less positive than for the natural uranium reactor. The principle reasons for this are 1) the lithium is more poisonous in the dry reactor and 2) the behavior of both the wet and dry reactivity with temperature and exposure is milder.

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DISCUSSION OF THE REACTIVITY CHANGES (CONT'D)

From the above considerations, it can be seen that the addition of both lithium and enriched uranium to the reactors lessens the control requirements compared to the normal natural uranium loading. However, in any nuclear safety analysis, particularly one involving the adequacy of the strength of the total control systems to hold the reactor subcritical under all circumstances (or allow a mild, self-quenched excursion in the melting reactor after all chances of restoring cooling are gone¹), one must consider all potential sources of large positive reactivity changes in the reactor. The positive reactivity sources are xenon decay, xenon escape from the reactor, physical removal of control elements and a possible positive moderator temperature coefficient at high exposures. In the E-N load, must be added the potential large reactivity gain resulting from losing lithium from the pile.

The preceding chemical and physical analyses have shown that there is a better chance that uranium will leave the reactor before the lithium. In addition, the analysis shows that a large rearrangement of both lithium and uranium occurs during the reactor meltdown. It is the purpose of the subsequent analysis to show that none of these changes can produce a supercritical state in the melting reactor.

We follow the same type of analysis as Brown did in HW-50284¹. We taken for our study case a startup from a minimum outage which can occur with essentially no horizontal control rods in the pile, no auxiliary poison charged, and low xenon backlog. This case of water loss during minimum startup represents the worst possible case from a total control standpoint. Our analysis assumes conditions which are somewhat extreme; the present reactor conditions preclude a critical state. Both the natural uranium solid and E-N I & E loading are considered.

The following assumptions are made:

1. The reactor has been down for 30 hours and has a minimum startup. The operating time after startup is very short -- just long enough to require almost total horizontal rod withdrawal. The graphite is thus still cold when water flow is completely and permanently lost.
- 2) The reactor is H reactor operating at 1900 MW. The ball 3X system capacity under these conditions is approximately 2500 ih. No power excursion results immediately after water loss. No additional controls can be inserted.
- 3) Xenon remaining in the pile is 750 in-hours. Neglect the small amounts of xenon and iodine produced during the short startup operation. Also neglect the small amount of iodine remaining from pre-shutdown equilibrium operation.
- 4) Reactivity jump due to water loss is assumed to be 800 ih in both the natural solid and E-N I & E loadings which is actually more than the measured cold reactor values. (See Table I).

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DISCUSSION OF THE REACTIVITY CHANGES (CONT'D)

- 5) Exposure is assumed to be 400 MWD/T average in the natural uranium pile and 500 MWD/T in the E-N pile. The net reactivity gain from the moderator temperature coefficient is $+2.5\% \Delta k$ for the natural uranium pile at the assumed exposure, but only $+0.5\% \Delta k$ for the E-N pile.
- 6) The metal coefficients are $-1.2 \text{ ih}/^\circ\text{C}$ and $-1.0 \text{ ih}/^\circ\text{C}$ for the natural solid and E-N I & E piles, respectively.
- 7) All tubes will be assumed to heat up as though they were central tubes. No account is taken of the fact that part of the tubes are lower power tubes and hence are slower to heat. The time required to reach melting temperature of uranium is 23 hours after loss of water.
- 8) The process tubes are blocked by UAl_3 formation in about 6 hours.

Figure 1 has been prepared to show the reactivity changes in the two piles during meltdown. The various portions of the figure may be described as follows:

- 1) Water flow is lost and the reactor scrams. The ball 3X safety system drops in and provides 2500 inhours of negative reactivity. Simultaneously, the water is expelled from the pile and a gain of 800 inhours results. Just after the shutdown the pile is thus 1700 inhours subcritical.
- 2) After shutdown, residual fission product heating raises the fuel element (natural uranium or E-metal) temperature. Since the fuel element is fairly well thermally insulated from the graphite the temperature increase is very small and hence the reactivity change is solely that from the negative Doppler coefficient. Only the E-N case curve is shown in the figure during this period, the natural uranium reactor would be driven further subcritical due to the more negative coefficient.
- 3) About six hours after the accident, the fuel element jacket temperature reaches $550 - 600^\circ \text{C}$ at which time the rapid diffusion of uranium into the aluminum begins. As postulated in the previous section, the diffusion is expected to penetrate into the N slug before enough heat is transferred to melt the lithium. In addition, the aluminum process tube will enter into the diffusion reaction also. and as the results of the KW incident showed, the volume expansion of the UAl_3 will effectively block the process channels, and the lithium will be either physically trapped or chemically bound within the matrix.

The effect on reactivity of the U-Al interdiffusion has been shown to be negative (HW-54188)* since the decrease in the resonance escape probability due to the U^{238} dilution more than offsets the increases in f and ϵ . Although, the net effect of the diffusion is negative, we shall ignore it in our analysis. An additional decrease in reactivity is introduced by the increased distribution of the lithium within the tube, and this is also ignored.

* In the analysis in HW-54188, the entire slug was assumed converted to the alloy. In our case, the net decrease in reactivity would not be as large as shown in the earlier analysis since the aluminum available is only sufficient to tie up 30% of the uranium, as UAl_3 . The KW incident showed UAl_3 to predominate.

FIGURE I

REACTIVITY TRANSIENTS DURING E-N AND NATURAL URANIUM PILE
MELTDOWN AT H REACTOR (UNIFORM LOADING)

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DISCUSSION OF THE REACTIVITY CHANGES (CONT'D)

- 4) Once good thermal contact is established between the heat source and the graphite via the UAl_3 filling the process channel, the graphite and metal temperature approach each other. Because of the large heat capacity of the graphite in the cell, the resultant temperature will be only a little above the original inlet water temperature. During the subsequent period then, the metal cools down and the decrease in reactivity from the metal coefficient is cancelled, as is shown in the curve in Figure I.*
- 5) The metal and graphite complex now heats up over a period of hours until at 23 hours after the accident the melting point of uranium ($1133^\circ C$) has been reached. During this period the lithium is either bound in the UAl_3 matrix which melts at $1350^\circ C$, or, if not, the blockage of flow-passages by the expanding UAl_3 makes it difficult to imagine much gross movement occurring. As evidenced from the KW incident, much of the uranium was forced by the UAl_3 pressure into cracks and crevices in the graphite. Physical rearrangement of the uranium in the cell causes a very large loss in reactivity and several experiments (See HW-50284) have been performed to indicate the order of magnitude of the loss. However, in our analysis complete credit for this loss is delayed until the melting of the UAl_3 occurs.
- 6) The heating of the graphite-uranium complex plus the continual decay of xenon produces the transients shown in Figure I between 7 and 23 hours after the loss of coolant. It is to be strongly noted that the net change in reactivity in the E-N case even at an average exposure of 500 MWD/T is negative, whereas in the normal loading the reactivity increases with time.
- 7) Eventually at a pile temperature of $1350^\circ C$, the UAl_3 melts and flows. The lithium, aluminum and uranium are now free to flow in the gravity field, and distribution of both uranium and lithium in the reactor lead to very large losses in reactivity.
- 8) The diffusion of lithium vapor, absorption in the graphite at temperatures above $1350^\circ C$ and the eventual boiling and distillation of lithium to colder regions of the pile at $2000^\circ C$ will also result in very large poisonous effects. Rather simple calculations have shown that in the N slug (in the dry pile) the lithium accounts for 14% in reactivity whereas, completely distributed in the graphite it is 35%, indicating in this extreme case that up to 65% of the lithium can leave the pile with no change in reactivity. Such a complete distribution is not physically possible, but sufficient lithium will be in regions of pile where its poison value is significantly greater. Since the uranium is much heavier than the lithium or aluminum, it will flow farther in the gravity field, and if passages to the exterior of the reactor are open, the material to come out of the reactor first will probably be the uranium.

* During this time some xenon has been decaying off, too, but for simplicity the gain in reactivity from this source is delayed in our analysis to be included in the much longer period from seven to twenty-three hours after the accident (See paragraph 6).

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CONCLUSIONS

The behavior of an E-N load will essentially duplicate the behavior of tube 4669 in the K west reactor incident. The introduction of N metal will not alter the situation until temperatures above 1350° C have been reached. In the interval from initial temperature to 1350° C, the KW reactor incident demonstrates quite precisely what will happen. Total obstruction of the coolant annulus by UAl₃ will occur when the uranium-AlSi interface has been at a temperature of 550° C to 600° C for two minutes or so. Pressures caused by the swelling UAl₃ will be great enough to extrude the uranium out of any holes and into the graphite weep holes and block interfaces. The key fact learned from tube 4669 KW is that the diffusion of uranium and resultant formation of solid, high melting UAl₃ occurs below the melting point of aluminum or lithium-aluminum alloy. Since the source of heat is the E metal it is probable, that any non-reacted uranium will melt, flow and react with any aluminum it contacts, before the heat from the E metal can cause melting of unreacted LiAl.

However, it is also possible for a fraction of the LiAl piece to melt without being tied up as spongy UAl₃. In this event, the melt would probably be trapped in situ between the walls of UAl₃ from the E piece, and the 72-S process tube or graphite if the tube has melted. In the event that the lithium metal does find a channel, the probability is very high that not more than one lattice unit of flow might occur before uranium was contacted. Most access volumes will be filled with cast uranium and/or UAl₃.

It is probable also, that the reaction of uranium on lithium-aluminum alloy will result in the formation of UAl₃ which will physically trap the lithium as part of the matrix. The resulting structure should securely bind the lithium in place.

The net result will be a more diffuse distribution of the E metal with possible impregnation of the graphite pores with molten aluminum or LiAl. The vapor pressure of the lithium will be markedly lowered by alloying, and its migration will not become appreciable until well over 2000° C. From a chemical viewpoint, at least, there appears to be no new hazard introduced with the LiAl metal than already exists in a normal loading of any Hanford reactor.

The nuclear physics of the E-N reactor is little different than that for the natural uranium reactor. Any differences are to the credit of the E-N reactor as far as nuclear safety is concerned. Smaller water-loss reactivity jumps, milder temperature coefficients, delayed influence on the dry moderator coefficient by plutonium buildup - all contribute to lessen the control requirements.

Chief concern in the E-N reactor is loss of lithium from the reactor. However, the expected chemical and physical changes wrought in the melting pile strongly point to the fact that the uranium and not the lithium will leave the reactor first if passage is available. Furthermore, the movement of both uranium and lithium from the fuel channel into a more diffuse distribution in the lattice cause very large losses in reactivity.

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It is thus the conclusion of this report that the introduction of lithium into a graphite moderated, water-cooled, uranium metal production reactor introduces no new problems in nuclear safety not already present in the natural uranium pile.

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