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FULL REACTOR-E-N-LOAD-REVIEW OF THE HAZARDS FROM A CHEMICAL VIEWPOINT

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FULL REACTOR-E-N-LOAD-REVIEW

OF THE HAZARDS FROM A CHEMICAL VIEWPOINT

by

W. R. DeHollander

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HANFORD LABORATORIES

By Authority of CG-PR-2

D.S. Lewis, 12-17-93.

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December 22, 1958

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- 2 -

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FULL REACTOR-E-N-LOAD-REVIEW
OF THE HAZARDS FROM A CHEMICAL VIEWPOINT

W. R. DeHollander

INTRODUCTION

Chemical Research and Development Operation was contacted by R. Nilson⁽¹⁾ of Irradiation Processing Department in order to obtain an analysis from a chemical viewpoint of the worst credible accident to a full E-N reactor loading. This analysis would be used to present to the ACRS directly or would be abstracted to prepare a supplementary report.

The primary purpose of this report is to present an analysis of the chemical behavior of an E-N loading in the event of the sudden loss of coolant to the entire reactor, with, however, normal operation of the control devices such that the reactor is initially driven sub-critical. We are here concerned with the effect of a rising temperature upon the E-N load, the rise in temperature being primarily caused by the release of fission product energy.

SUMMARY AND CONCLUSIONS

The behavior of an E-N load will essentially duplicate the behavior of tube 4669 in the K West reactor incident, reference documents HW-35821⁽²⁾ and HW-35822.⁽³⁾ The introduction of the 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 will occur when the uranium-AlSi interface has been at a temperature of 550 C to 600 C for two minutes or so. At this time rapid diffusion of uranium into the aluminum and/or LiAl of the N piece will create voluminous UAl_3 , an intermetallic compound whose melting point is about 1350 C. The resultant swelling will completely block the coolant annulus, and the 72-S process tube will become one with the slugs. Pressures caused by the swelling UAl_3 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 UAl_3 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 UAl_3 . In this event, the melt would probably be trapped in situ between walls of UAl_3 from the E piece, and the 72-S process tube or graphite if the tube has melted. Very little if any gross flow of LiAl should occur. In the event that the 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 UAl_3 .

It is probable also, that the reaction of uranium on lithium-aluminum alloy will result in the formation of UAl_3 which will physically trap the lithium as part of the matrix. The resulting structure should securely bind the lithium in place.

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HW-58813

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.

DISCUSSION

In such 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 (EW-35821)(2) and EW-35822(3)). 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 happened were beautifully done and 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.

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- 5 -

HW-58813

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,^(4,5) 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.

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 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 C. Therefore, at 1400 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 illdefined upper temperature in the neighborhood of 2000 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.⁽⁶⁾ Around 2000 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 C. These slugs will also start to swell at this time and might add to the blockage of the channel which

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EW-58813

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 $-50^\circ C$ 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.

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 .

An experiment to answer the above question might be fruitful of results.

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