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Division of Union Carbide Corporation

Post Office Box X
Oak Ridge, Tennessee

External Transmittal
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ORNL
CENTRAL FILES NUMBER

58-8-2

DATE: August 1, 1958
SUBJECT: Trip to ALCO Products, KAPL, Combustion Engineering Co. and Ft. Belvoir (APPR): Re: Cooling Water Activity Buildup in Pressurized Water Reactors
TO: Listed Distribution
FROM: H. N. Culver

COPY NO. 29

Abstract

The activity in the cooling water and in the demineralizer of the NMSR has been examined and reported in CF-58-5-83. This study established the fact that there were several uncertainties associated with predicting activity levels in water-cooled reactors. In order to evaluate these uncertainties, discussions were held with personnel at KAPL, ALCO Products, Combustion Engineering, and Ft. Belvoir.

The topics discussed and covered in this memo are:

- a. Effect of in-pile and out-of-pile material on water activity buildup.
- b. Effectiveness of the purification system in limiting activity buildup.
- c. Factors influencing deposition.
- d. Importance of n, p reactions.
- e. Importance of water impurities.
- f. Fission products in the coolant.
- g. Dose levels inside the reactor compartment.

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As part of the ORNL effort in support of the NMSR, studies have been made to establish what levels of activity might possibly exist in the cooling water and in the demineralizer of the N. S. Savannah. (Ref. 1.) The results of these studies were obtained by assuming several conditions in the system which are not clearly understood at this time, but which were necessary in order to obtain analytical results. In order to determine if better methods exist for solving such a problem or if better assumptions could be made, discussions were held with the following people at the various locations indicated:

ALCO Products	Bill Brown J. Zegger
KAPL	David Adams Sherman Williams
Combustion Engineering Co.	Jerry Davis
APPR - Ft. Belvoir	Jim Leslie Lew Heider

The ORNL Chemical Technology Division, which has an interest in waste disposal and the purification system for the NMSR, was represented by Mr. J. T. Roberts. A report covering this part of the discussions has been made in CF-58-7-109. The following report covers the general information having to do with activity buildup in water systems.

The calculation of the cooling water activity in Ref. 1 had been made based on the following main assumptions:

- 1) Corrosion products formed in out-of-pile regions are not deposited in the core and then released. Out-of-pile material is only activated as it passes through the core regions.
- 2) Material which is released from the primary loop surfaces (both radioactive and nonradioactive material) enters the water and remains in the water until it is removed by either decay or by purification. This implies that the purification system is 100% effective and that there is no deposition of nuclides on primary loop surfaces.

Both of these assumptions were made so that a solution would be possible. However, it was recognized that such factors as the in-pile vs out-of-pile material, deposition, and effectiveness of the purification

system could alter the resulting water activities by several orders of magnitude.

Factors Influencing the Buildup of Activity in Water Systems

1. In-Pile vs Out-of-Pile Material

The importance of activity buildup from out-of-pile material is largely a function of the materials used for construction in the system. If a system has zirconium-clad fuel elements and stainless steel for structural materials in the external loop, the out-of-pile material will be the greatest contributor to the activity. However, if a system has stainless steel for both the fuel element clad and for the piping in the primary loop, most of the activity will be due to the activation of core material only. If the stainless steel in the core has a lower cobalt content than the stainless steel in the primary loop piping and equipment, the activity resulting will be associated with the in-pile material, but it is not clear if the reduced cobalt content will give a proportional decrease in activity levels. The factor of importance is the deposition and release rates associated with the materials in pile and out of pile. It now seems that the deposition and release rates for stainless steel on zirconium are very much greater than for stainless steel on stainless steel. Therefore, out-of-pile material is more important only for fuel elements with zirconium clad. The information obtained from the APPR with Core II will determine if reducing the cobalt content of in-pile material has a pronounced effect on overall activity buildup.

2. Effectiveness of the Purification System

The real importance of the purification system for reducing activity is directly related to the other processes for removal of activity, the deposition rate, and the decay of radioactive nuclides. The purification system reduces the impurity level in the coolant, and since the mechanism for deposition depends upon the transport of nuclides, the purification system rate would decrease the amounts capable of being deposited. Although the importance of the purification system is decreased by deposition, the fact that activity builds

up to high levels in the demineralizer indicates that it is reducing the total activity in the primary system, and is therefore of importance in reducing the overall activity.

3. Factors Influencing Deposition

In calculating activity buildup on the surfaces of piping and equipment, it is important to know the deposition rate for the individual nuclides as well as corrosion rates and release rates. Since these rates are very difficult to measure experimentally, values have been arrived at which are consistent with the experimental data and which satisfy theoretical calculations. Since it is difficult to predict release rates or deposition rates for systems lacking experimental information, it is necessary to assume that the deposition of radioactive nuclides occurs uniformly over the primary surfaces. Such an assumption results in a deposit of a particular number of mg/cm^2 of primary surface and assumes that the composition of the deposit is the same as for the material being corroded. This then assumes that factors such as velocity and temperature do not induce a preferential deposition. Since experimental results indicate that activity deposition is higher at the inlet to the steam generators than at the outlet, there is sufficient indication to believe that deposition is a function of temperature. The evidence that activity builds up to high levels wherever low velocity or dead legs occur indicates the effect of velocity on activity due to deposition. Since essentially all the activity in a section of piping is associated with the activity deposited on surfaces, rather than either in solution in the water or being carried in the water, it might be expected that the real effectiveness of the purification system is greatly overestimated by assuming all the system activity to be in the water.

4. Importance of n, p Reactions

The most important n, p reaction is the reaction on nickel producing Co^{58} . The real importance of this reaction depends upon the fast flux in the reactor system since it is a process involving fast neutrons. Therefore, in a reactor system the ratio of fast to thermal flux, as well as the cobalt content, will determine the relative importance of Co^{58} and Co^{60} .

5. Importance of Impurities in the Water

The prediction of activities in the NMSR had indicated that Na^{24} would result in the highest activity immediately after shut-down. This was based on an assumed impurity level of 0.1 ppm of Na^{23} . Although the data from the APPR have not shown sodium activity to be of importance, the actual amount of activity from sodium is not known since the data obtained have been only for long-lived nuclides. Future programs will determine the actual amount of activity from this source. One of the more important activities which may be introduced into a water system is the activity associated with argon. Since this is the only gaseous activity to contend with, assuming no fission product releases, the level should be controlled by using deaerated water. The nuclide K^{38} , which was included in the B&W report but was not included in Ref. 1, is not found in the APPR, but here again the APPR data measured only long-lived activities. None of the people contacted feels that K^{38} is the source of the activity attributed to it.

6. Fission Products in the Coolant

The buildup of activity on surfaces will come from two sources; 1) the normal sources including corrosion products and water impurities, and 2) the fission products which are released under abnormal circumstances. Since the fuel element for the NMSR is of a different material from any of the other water reactors presently in operation, a direct comparison is not possible. The naval reactors have been designed for service assuming no release of fission products. Therefore, if the naval reactor fuel elements were to rupture or to leak appreciable amounts of fission products, the system would be shut down. Fission products have been detected in the APPR; however, the level of activity has been so low as to present no problem of operation. It has not yet been determined if the activity is from a small defect, surface contamination, or diffusion through the clad. Reference 1 has attempted to predict the levels of activity in the demineralizer and in the water for an assumed number of defects. These results have been obtained by assuming the same mechanisms for removal as for the corrosion products, namely decay and purification. As indicated in

items 1-3, the activity in the primary system will probably be increased since the activity is not so much in the water as on the surfaces, and therefore the purification system is not as effective.

7. Doses Inside the Containment Vessel

The level of radiation inside the reactor compartment will come from four primary sources after shutdown; 1) the radiation from fuel element fission product decay and structural material activation which penetrates the primary shield, 2) primary piping and equipment which has been activated by either reactor neutron leakage or by neutrons being circulated in the coolant, 3) corrosion products and impurity activation deposited on the surfaces of the primary system, and 4) fission products deposited on the primary surfaces. The relative contribution of each of these sources to the total dose inside the reactor compartment or containment vessel will depend to a large extent upon the types of fuel element and its failure or defect rate, the amount of primary shielding, the type of structural materials used in the primary system, the effectiveness of the purification system, the conditions under which the primary system operates, and the flux level or power level of the reactor.

The experience with operating reactors indicates that the contribution from activated primary piping and equipment is negligible compared to the other sources. The doses inside the reactor compartment of the APPR are in general relatively low. The general background activity 30 min after shutdown is about 50 mr/hr. However, there are areas where the doses are appreciably higher; for example, the dose adjacent to the heat exchanger is 440 mr/hr 30 min after shutdown, and the dose at the reactor outlet 30 min after shutdown is about 380-440 mr/hr. At various locations where deposition or crud collection is highest the dose levels are very much higher, also as one would expect. At the pressurizer leg the dose is as high as 1.8 r/hr and adjacent to the flow tubes (dead legs to instruments) the dose is about 940 mr/hr. These doses are primarily associated with the activated crud deposits on the surfaces of the equipment due to corrosion products.

The NMSR activity levels in general will be similar to the levels found in the APPR except for the difference in power levels. There are a number of other factors, however, that may tend to increase the dose levels in the NMSR above those indicated above:

- a) The main contribution to the dose from deposited activity on piping is due to Co^{60} . Since the APPR has not operated long enough for the Co^{60} to reach equilibrium, the doses from this source should increase as a function of time.
- b) The dose from Co^{60} will depend upon the concentration of Co^{59} in the core material. Since this has not been specified for the NMSR, the amount of activity in the NMSR, as compared to the APPR, may be either appreciably greater or less.
- c) The purification rates for the two systems are not the same. Since the real effectiveness of the purification system is not known, the effect on the doses is also unknown.
- d) The physical layout of the piping and equipment will determine to some extent what levels of activity will be present in the NMSR. Elimination of dead legs or lines where crud is able to collect will greatly reduce the dose from activated corrosion products.
- e) The NMSR will probably operate with defective fuel elements. This will add an additional source of activity in the reactor compartment which is not expected in the APPR or the naval reactors. The actual contribution of this source will depend upon the number of defective fuel elements, the amount of cleanup in the system after numerous defects, and the actual size of the defect (i.e., is it a 5-mil defect or a rupture).

In addition to the above items influencing the actual dose in the reactor compartment, the amount of primary shielding will influence the total dose at a particular place inside the containment vessel.

In an attempt to predict the levels of activity in reactor systems, KAPL and Combustion Engineering have developed elaborate equations (Refs. 2-5) similar to those in Ref. 1 but containing terms to

take into account the corrosion of out-of-pile material, deposition in the core, and then release after activation. These equations have been developed based on experimental results and have therefore been able to predict to a greater degree of accuracy the levels of activity present in their systems. Such a solution to the activity problem is a more elaborate method of predicting activity than the method of Ref. 1, but it is probably a necessity for systems having Zr clad since a) the deposition of stainless steel on Zr is much greater than stainless steel on stainless steel, and b) the activation of Zr in the core is very much less than the activation of stainless steel in the core. In the case of all-stainless steel systems, it is felt that using the method of Ref. 1, the levels of activity in the primary system are predicted quite well since most of the activity buildup is due only to in-pile regions. However, the fact that nuclides do not remain in the water but are deposited on the surfaces in the primary system tends to overestimate the amount of activity in the demineralizer and underestimate the activity remaining in the system. Since the equations of Ref. 1 were written to take into account the deposition of activity, these equations should be solved using deposition rates which would represent an all-stainless steel system. The deposition rate used should be such that the resulting activity on the pipe surfaces should greatly exceed the activity in the water, the exact ratio having been found experimentally. Such a solution would decrease the amount of activity in the demineralizer for the long-lived nuclides. It is felt that it would be possible to ascertain deposition rates for the longer lived nuclides by varying the purification rate and recording the deposited activity on a given length of piping. Having established a uniform deposition in the primary loop, it would then be possible to predict activity levels at various places in the reactor compartment. Assuming a uniform distribution of these nuclides would certainly introduce an additional error since it is well known, as was previously indicated, that both temperature and velocity influence the buildup of deposited or collected activity. However, having established a uniform activity on surfaces it

would then be possible to postulate certain areas of higher concentration and thus predict higher levels at these points based on APPR data.

References

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2. SIC/FP/56/103.
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