SURVEY
OF
FISSION- AND CORROSION-PRODUCT ACTIVITY
IN
SODIUM- OR NaK-COOLED REACTORS

By

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ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN ROCKWELL CORPORATION

CONTRACT: AT(04-3)-701
ISSUED: FEBRUARY 28, 1969

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I. INTRODUCTION

While there are a number of reported instances of fission-product release to the coolant and cover gas of sodium- or NaK-cooled reactors, these occurrences were incidental to the pursuit of some other reactor operational objective. As a consequence, data on fission-product distributions, and the resulting residual radiation levels in the affected systems, are generally sketchy, and are only found in widely scattered references. While nothing can now be done about the limited quantity of data reported, these data have been assembled and summarized, and are presented in this report. From this summary, it is possible to discern a general pattern of common behavior and certain notable exceptions. These are discussed in the next section.

II. DISCUSSION

The summary of data for seven sodium- or NaK-cooled reactors is presented in Table 1.

Discussion of the data for each reactor is given here. In examining Table 1, it should be noted that the absence of data in a given block does not necessarily imply a negative finding; rather, it merely means that no data were reported (Note: The bibliography, at the end of this report, summarizes the reference information for each reactor).

A. SRE

Most of the data in Table 1 for the SRE are based on samples and measurements obtained during the three months immediately following the fuel failure incident of July 26, 1959. As indicated, however, some of the data were obtained as much as five years later.

As is well known, the cause of the SRE fuel failure was the blockage of coolant channels by particulate carbonaceous decomposition products of Tetralin which had leaked into the sodium coolant, or by a combination of these carbonaceous particulates with sodium oxide. While this caused damage to 13 of the 43 fuel elements in the core, and hence produced a substantial fission-product release, there is evidence to indicate that the presence of the carbon substantially modified the behavior of the fission products in the coolant. Thus, the SRE experience is believed to be not necessarily typical of fission-product release in sodium systems in the absence of a high-concentration carbon impurity. This atypical behavior is ascribed to the "gettering" of fission products by the carbon, and is illustrated by the data shown in Table 2.

The activity of carbonaceous material shown in Table 2 is the activity measured in the suspended carbonaceous material which had been removed from the sodium by filtration. The activity of sodium given in Table 2 is the activity of the filtrate.

The observed $^{95}\text{Zr-Nb}$ activity reported in both Tables 1 and 2 is also worthy of note; in that, while it may be due to released fission products, it may also be due to the activation of the ^{94}Zr present in the SRE zirconium

	SRE ^c	HNPF	Fermi	BR-5	Dounreay	EBR-2	S8ER
Maximum Power (Mwt)	20	254	100	5	60	62.5	0.6
Energy Generated (Mwd)	2425.8	26,615	855	1625	22,500	Not applicable	212
Fuel	Unalloyed uranium metal + experimental elements	U - 10 wt % Mo	U - 10 wt % Mo, Zr cladding	PuO ₂	U - 9 wt % Mo	U-Mo	(U-Zr)H _x
Coolant	Na	Na	Na	Na	NaK	Na	NaK
Neutron Spectrum	Thermal	Thermal	Fast	Fast	Fast	Fast	Thermal
Typical Reactor Outlet Temperature (°F)	1000	950	800	930	445	900	1300
F. P. ^a Released (Ci)	5,000-10,000	No failed fuel elements	10,000	Sustained operations with known defected fuel elements	Open fuel elements	Failed fuel irradiation capsule	Many failed fuel elements
F. P. Found in Coolant	¹⁴¹ Ce, ¹³¹ I, ¹⁰³ Ru, ¹³⁷ Cs, ⁸⁹ Sr, ⁹⁰ Sr, ⁹⁵ Zr-Nb, ^d ¹⁴⁰ Ba-La, ¹⁴⁴ Ce		¹⁴⁰ Ba-La, ¹³⁷ Cs, ⁸⁹ Sr, ¹³¹ I	¹³⁷ Cs, ⁹⁵ Zr-Nb, ¹⁴⁰ Ba-La	¹⁴⁰ Ba-La, ¹⁴¹ Ce, ¹³¹ I	¹³⁷ Cs, ¹³¹ I, ¹³³ I	¹³⁷ Cs, ¹³¹ I, ¹³² Te-I, ¹²⁵ Sb, ⁸⁶ Rb, ¹³⁴ Cs ^j
F. P. Found on Primary Pipes	⁸⁹ Sr, ⁹⁰ Sr, ⁹⁵ Zr-Nb, ^d ¹⁴⁴ Ce, ¹³⁷ Cs, ¹⁰⁶ Ru		¹⁴¹ Ce, ¹⁴⁴ Ce, ¹³³ I, ¹⁰³ Ru, ⁹⁵ Zr-Nb ^g	¹³⁷ Cs, ⁹⁵ Zr-Nb ⁱ			⁸⁹ Sr, ⁹⁰ Sr, ⁹⁵ Zr-Nb, ¹⁰³ Ru, ¹⁴⁴ Ce, ¹⁰⁶ Ru-Rh, ¹²⁵ Sb, ¹⁴⁰ Ba-La, ¹⁴¹ Ce
F. P. Found in Cover Gas	¹³³ Xe, ⁸⁵ Kr	¹³³ Xe, ¹³⁵ Xe, ⁸⁵ Kr	¹³³ Xe, ¹³⁵ Xe, ⁸⁵ Kr	¹³³ Xe, ¹³⁵ Xe, ⁸⁵ Kr	¹³³ Xe, ¹³⁵ Xe, ⁸⁸ Rb, ¹³⁸ Cs	¹³³ Xe, ¹³⁵ Xe	¹³⁵ Xe, ³⁹ A ^k
F. P. Found in Cold Trap	¹³⁷ Cs, ¹²⁵ Sb ^e			¹³⁷ Cs			No preferential concentration of F. P. observed in NaK obtained from cold trap
C. P. ^a Found in Coolant	⁵¹ Cr, ⁹⁵ Zr-Nb, ^d ⁵⁴ Mn, ⁶⁰ Co, ⁵⁹ Fe, ¹²⁵ Sb						⁵⁶ Mn, ⁶⁰ Co
C. P. Found on Primary Pipes	⁹⁵ Zr-Nb, ^d ⁵⁴ Mn, ⁶⁰ Co, ⁵⁹ Fe	⁵⁴ Mn, ⁶⁰ Co					⁵⁸ Co, ⁶⁰ Co, ⁵⁴ Mn, ⁵⁹ Fe
C. P. Found in Cold Trap							
Highest Residual Radiation Level Observed ^b							
Primary Piping	3 R/hr	50 mr/hr	3 mr/hr (section of pipe stored in fuel transfer pot read 180 mr/hr at 0.5 in.)	General Comment: <10 R/hr (reduced to 36 mr/hr after extensive decontamination)	General Comment: Can be decontaminated by acid pickling, down to 200 mr/hr		
Intermediate Heat Exchanger	11 R/hr ^f	12 mr/hr	6 mr/hr				
Cold Trap	81 R/hr	50 mr/hr	40 mr/hr ^h				

Notes: a F. P. = fission products, C. P. = activated corrosion products

b After decay of ²⁴Na

c SRE data must be interpreted in light of the presence of large quantities of carbonaceous materials in the coolant, and use of zirconium moderator cans.

d ⁹⁵Zr-Nb may be activated corrosion product from zirconium moderator cans.

e Measured in 1963

f Measured in 1964, after removal of shell, looking down cold-leg tubes

g ⁹⁵Zr-Nb may be activated corrosion product from zirconium fuel cladding.

h Cold trap operation terminated 2 hr after shutdown

i ~90% of activity on pipe wall

j The activities due to ⁸⁶Rb and ¹³⁴Cs are attributed to activation of impurities (⁸⁵Rb and ¹³³Cs) in the coolant.

k ³⁹A activity from reaction, ³⁹K (n,p) ³⁹A

Table 1. Summary of Fission- and Corrosion-Product Distribution Data for Sodium- or NaK-Cooled Reactors

AI-AEC-MEMO-12790

TABLE 2
RADIOACTIVITY ANALYSIS OF TYPICAL SAMPLE OF
CARBONACEOUS MATERIAL FROM
PRIMARY SODIUM SAMPLE

Isotope	Activity of Carbonaceous Material* (μ Ci/gm of particles) [†]	Activity of Sodium (μ Ci/gm Na) [†]
¹³⁷ Cs	1.1×10^3	0.45
¹³⁴ Cs	2.9×10^1	0.006
¹⁴¹ Ce	2.3×10^2	0.0007
¹⁴⁴ Ce	2.9×10^2	0.00039
⁹⁵ Zr-Nb	2.2×10^2	0.018

*Obtained from reactor on 10/31/59

[†]As of 7/26/59

moderator cans and subsequent corrosion. If this latter mechanism is predominant, this activity would only be observed in reactors utilizing zirconium-containing components.

Comparison of SRE piping, intermediate heat exchanger (IHX), and cold trap residual radiation levels with those listed for Fermi indicates the SRE levels were $\sim 10^3$ higher for approximately the same fission-product release. A possible explanation for this situation is the gettering of fission products by the carbon in the SRE system, combined with the deposition of the carbon on the walls of the primary system piping, IHX, and cold trap. This carbon deposit was sufficiently thick, in one instance, to plug a number of tubes at the end of the cold leg of the SRE IHX. It was these deposits that gave rise to the 11 R/hr radiation level (measured in 1964) quoted for the IHX in Table 1. Further discussion of the differences in fission- and corrosion-product behavior between SRE and Fermi is given in the section on the Fermi reactor.

B. HNPF

While the gaseous fission products shown in Table 1 were observed in the Hallam reactor, limited radiometric analysis of the coolant did not reveal the

presence of any other fission products. Analysis, in 1968, of stainless steel samples from the Hallam primary system did reveal the presence of the activated corrosion products, ^{54}Mn and ^{60}Co , diffused into the surface of the steel. Because there was not significant solid fission-product release from the Hallam fuel, the observed residual radiation levels are attributed to activated corrosion products, activated coolant impurities, and ^{22}Na .

A potentially important problem was discovered in connection with Hallam operation and retirement (viz, certain components could not be completely drained of sodium). Long-lived activity in the remaining sodium therefore added to the radiation levels encountered during access to such components.

C. FERMI

The most notable feature of the Fermi data is the large discrepancy between the radiation levels measured at Fermi and those measured at the SRE. There are several factors which contribute to a partial explanation:

- 1) The energy generated by Fermi was about one-third of that generated by the SRE, and it was generated over a much shorter time. This would favor the buildup and rapid decay of the shorter-lived isotopes.
- 2) At the time of the Fermi incident, the reactor was approaching power after having been essentially shut down for over a month (integrated thermal power was 1 Mwd in September and another 1 Mwd in October, during approach to power). The SRE incident occurred at the conclusion of a 16-Mwd run; this run, however, also followed a shutdown period of ~ 1 -month duration.
- 3) Because the oxide concentration in Fermi was maintained at a very low level, and because the sodium outlet temperature was kept below 700°F , it is probable that very little corrosion (and hence, generation of corrosion products) occurred in the primary system. In the SRE, reactor outlet temperatures of 1000°F or above had been achieved for sustained periods of operation. Further, although sodium temperatures were somewhat lower for the run which terminated in the SRE fuel-melting incident, this run was characterized by very high sodium oxide concentrations (plugging temperature as

high as 455°F, at one point). Thus, high temperatures and high oxide concentrations in the SRE would probably result in substantial generation of corrosion products.

- 4) As regards radiation levels in the cold traps, the SRE cold trap was placed in operation 17 hr before reactor shutdown, and maintained in continuous operation for an extended period of time thereafter. In the case of Fermi, the cold trap was operating at the time of the incident, and was turned off about 2 hr after reactor shutdown. In the postincident period, the Fermi cold trap was operated for only 8 hr on each of two different occasions, for the purpose of taking coolant samples.
- 5) The presence of large quantities of carbon in the SRE may have caused a different distribution of fission products in that reactor than in Fermi. Carbon appeared to be universally dispersed throughout the primary loop of the SRE; and, acting as a getter, the carbon could have produced a similar distribution of radioactivity. In the absence of carbon, geometrical factors could have played a more important role. In Fermi, with a larger upper sodium pool than the SRE and with a contiguous fuel transfer pot, there would appear to be more opportunity for fission products to deposit in the core vessel and fuel transfer pot. As shown in Table 1, a section of pipe removed from the fuel transfer pot, 3 months after the incident, exhibited a radiation level of 180 mr/hr at a distance of 1/2 in. Indeed, if the fuel transfer pot was maintained at a lower temperature than the upper sodium pool, it is possible that the surfaces in the fuel transfer pot acted as a sink for deposition of fission products.

As a matter of similarity with the SRE, the ^{95}Zr -Nb activity observed in Fermi may have been due predominantly to corrosion of the zirconium fuel cladding.

Finally, while only low levels of residual radioactivity were measured at Fermi, the operating staff reports that studies indicate plate-out losses from the coolant may be as large as 93% for ^{89}Sr and ^{90}Sr , 8% for ^{137}Cs , and 75 to 84% for ^{140}Ba -La.

D. BR-5

The data pertaining to the Russian BR-5 reactor are of particular interest, since this is a PuO_2 -fueled, sodium-cooled reactor which was intentionally operated for an extended period of time following fuel failure. The initial evidence of fuel failure was the detection of ^{133}Xe in the cover gas when the plutonium burnup reached 2.4%. Solid fission product (^{137}Cs) escape was indicated when the plutonium burnup reached 3.2%. Operation was continued until a burnup of 5% was achieved, at which time operations were terminated because of failure of a primary pump. After decay of the ^{24}Na activity, draining the primary system did not reduce the residual radiation levels, thus indicating that most of the activity was retained on the pipe walls.

At this point, the primary system was partially decontaminated by treating with steam, followed by several cycles of successively filling the system with 0.5% KMnO_4 solution, followed by a mixture of 5% nitric acid and 1% oxalic acid, and finally followed by "distillate" (at first, I thought "distillate" meant kerosene; but on further reading, I suspect it really means distilled water). The principal contamination found in the rinse solutions were ^{137}Cs and $^{95}\text{Zr-Nb}$.

The core was then reloaded with 63 of the old plutonium fuel elements and 50 new elements, and operation was resumed.

A number of tests were conducted to determine the effect of operating conditions on fission-product concentrations in the coolant. In general, fission-product concentrations were found to be independent of coolant temperature; but they were dependent on reactor power, increasing with increased power, presumably due to a greater rate of generation and release. Fission product concentrations in the coolant (particularly, ^{137}Cs) were also found to increase substantially with increases in cold trap temperature.*

The Russians conclude that "the presence of solid and gaseous fuel fission products in the coolant and gas of the primary circuit has produced no special complications of the reactor operation (sic)." They also conclude that

*This is in qualitative agreement with cesium behavior observed in current studies at Atomics International.

"sodium-oxide cold traps are also traps for catching solid fuel fission products." I do not know whether we can agree with their first conclusion, on a philosophical basis; and I have certain technical reservations about their second conclusion, because their data is so heavily weighted by observations of ^{137}Cs behavior. Nevertheless, the basic BR-5 data are probably the most pertinent presently available, as regards LMFBR operation with defected fuel elements.

E. DOUNREAY

The data for the Dounreay Fast Reactor (DFR) reflect the behavior of a fast reactor employing open fuel (i. e., coolant flows both inside and outside the cladding). The data are not too directly applicable to an LMFBR, because of the U-Mo fuel and the very low NaK primary coolant temperature (445°F). Because of this low temperature, little corrosion would be expected, and no data on activated corrosion products were reported. A large quantity of fission products apparently escaped from the fuel; however, the only information obtained concerning radiation levels is that given in Table 1.

F. EBR-2

As indicated in Table 1, the only data on fission and corrosion products in the EBR-2 are those derived from the failure of a fuel irradiation capsule. A pinhole failure is indicated, since only gases and volatiles were observed.

G. S8ER

Operation of the SNAP 8 Experimental Reactor (S8ER) was characterized by substantial release of fission products due to cladding failure occurring in about 80% of the fuel rods during the reactor's extended endurance run, and by the uniquely high coolant temperature. While there are significant differences between the S8ER and conventional power reactors, as regards fuel and coolant materials, examination of the data shows that the fission- and corrosion-product occurrence and distribution is similar to that observed in the more conventional power reactors. The principal exception is the occurrence of ^{39}A in the cover gas, arising from the $^{39}\text{K} (n,p) ^{39}\text{A}$ reaction which occurs with the potassium fraction of the NaK coolant.

III. CONCLUSIONS

In the event of significant fuel cladding failure, the following fission products will probably be present at the locations indicated:

Cover gas:	^{85}Kr , ^{133}Xe , and ^{135}Xe
Coolant:	^{131}I , $^{140}\text{Ba-La}$, $^{95}\text{Zr-Nb}$,* and ^{137}Cs
Primary system walls:	$^{95}\text{Zr-Nb}$,* ^{141}Ce , ^{144}Ce , ^{89}Sr , ^{90}Sr , ^{137}Cs , ^{106}Ru , and $^{140}\text{Ba-La}$.

Where coolant temperatures are 1000°F or above, corrosion of core structural and cladding materials is likely to occur. Some of this material will be radioactive, and will deposit on the walls of the primary system. The principal isotopes which will be found are ^{54}Mn and ^{60}Co .

The effectiveness of cold traps in the removal of fission and corrosion products other than ^{137}Cs remains to be determined, for systems not containing large quantities of carbonaceous materials.

*Assuming $^{95}\text{Zr-Nb}$ is a fission product, not an activated corrosion product.

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