OBJECTIVE

In normal operation of LMFBR, neutron-induced radioactive impurities released from composed material of the core, system components, and itself are introduced to primary coolant system, where any effective radiation shields and trapping devices for radioactive materials are hardly installed.

For the purpose of simplification of this problem, the radionuclides in question may be classified into two categories as major possible source of radiation hazard in the system maintenance and threat of release to environment. The one sort of nuclides is corrosion products leached from the core, and the other is tritium which is released as well to the coolant system through cladding material in the core with diffusion.

As to corrosion product nuclides, experimental research work on the behavior of these materials released in flowing sodium at O-arai Engineering Center is carried out for assessment of accessibility of the crews to the system piping and components. Several test runs, in correlation with the corrosion study of cladding material, have been performed to observe the pattern of
transport and deposition of corrosion products in Activated Material Test Loop-I.

EXPERIMENTAL PROCEDURE

An irradiated SUS316 specimen, a source of corrosion products which had a total activity of 0.5 Ci (Table 1) has exposed to flowing sodium at the maximum temperature region of the test loop so as to represent the behavior of corrosion products in primary circuits of LMFBR's.

The test loop (Fig. 1) of 25 kg sodium inventory, is composed of monometal SUS316 for the mass transfer analysis. This forced-convected sodium loop has two systems of a main test and a purification circuit which are circulated by an electromagnetic pump.

A 750 hr-test run was carried out subsequent to the three runs for preliminary test without any radioactive specimen.

Analytical method are summarized as follows:

during the loop operation

On-line γ-ray measurement, sighting two particular regions of the loop, one of which is hot leg located at 150 cm downstream distance from the activated source specimen, and the other is cold trap with the filler of stainless mesh having effective surface area equivalent to the wall of the circuit tubing (3/8 inches in diameter) of 6 m in length.

Chemical analysis and radioactivity measurement of circulating sodium sample taken from the purification circuit.

before and after the operation

Weighing, metallurgical examination, and measurement of specific or deposited radioactivity of SUS316 specimens exposed to sodium flow at hot and cold legs.

on completion of a series of the operation

Radioactivity measurement of tubular sample cut out from the entirely disassembled test loop.

RESULTS AND DISCUSSION
corrosion and leaching

(1) Estimating from the deposition rate of corrosion products, the initial leaching rate of activated specimen does not so accurately agree with any of such experimental expressions which so far have been reported. In this experiment, especially with regard to Cr, there has occurred a certain constant rate of leaching during about 10 days successively from the beginning of the experiment. After this period, the leaching rate rapidly declined. (Fig. 2)

It is assumed that this initial leaching rate has a close relation with initial impurity concentration in sodium, exposed surface area of tubing and components, sodium inventory, and also temperature gradient of the loop.

(2) Although it is not clear whether it is decidedly called the initial leaching transient effect, the following selectivity of leaching roughly could be given relating to some nuclides.

\[ Mn > Cr > Co \]

Particularly, selective leaching of Mn > Co is substantially clear.

However, the data are not obtained sufficient to compare that of Mn between Fe. (Figs. 2, 3, 4 and Photo. 1)

As for Zn, though the extent of leaching is not clear, it is showed to leach remarkably comparing with other major metallic elements.

(3) The relation between the performance of the cold trap and the corrosion rate has been investigated. It is found out that the corrosion rate does not slow down without the cold trapping, and quite contrary to this, it would tend to rather accelerate leaching of Cr and Co.

This fact very well endorses that the corrosion area for mass transfer in sodium system is the gradient of the concentration of alloy constituent elements in sodium. (Figs. 2, 3)
In normal operating condition that temperature of the cold trap is made to be lowered, oxygen impurity level in sodium also tends to decline and then as a result corrosion rate would be generally restrained. However, if the cold trap would be expected to remove effectively even these corrosion products as well as oxygen, and as the result, to lower the concentration of these products in the entire system, then considerably different aspect of material leaching and transfer might be apparent. If the oxygen concentration is sustained within a certain constant level, it is probable that the leaching rate of high temperature region would well be advanced when the cold trap is kept on operating through the experiment.

This is because, in the region where the concentration of corrosion products in sodium is low, solubility of these nuclides might be enhanced. While on the contrary, when the concentration has reached its saturation level, leaching probably would not possibly occur even if in the region of high temperature region. The fact that there was provided in this loop at the upper stream of an activated specimen a cartridge heater (though the flow velocity in it was negligible small) which would form a corrosion region having a further large exposed area and exhibiting higher temperature. In addition to this situation, it is necessary to be taken in account that relatively high flow rate in the cold trap was kept on during the experiment (the ratio of it to main flow is 1:3), when estimating a cause for such a unusual indication of the phenomena above stated.

These results, accordingly, can not be applied readily to the actual coolant system of reactor. However, the experiment carried out using AMTL-l for comprehensive understanding relating to corrosion and mass transfer demonstrates that unless many essential parameters not only flow velocity, temperature, and oxygen impurity level in sodium, but also operating conditions in the low temperature region, specimen's position, and the thermal flow in the system are correctly defined, there would be possibility of obtaining insufficient experimental data relating to this research.

(4) The cold trap temperature was sustained at 200 °C during the major part of the loop operation period, and the oxygen concentration in sodium was determined to be about 12 ppm by a chemical analysis of a sodium sample taken from expansion tank of the loop during its operation. This shows a fairly good agreement with the oxygen solubility curves recommended by Eichelberger, and Noden.

(5) At the high temperature region of 650 °C about 50 cm downstream from a main heater of the loop, an irradiated (by the flux of thermal neutron 4 x 10^20 n/cm², fast neutron 1MeV 10^20 n/cm²) trial-made cladding tube specimen for "JOYO" was exposed for 750 hours into the sodium flow of 2.5 m/sec velocity, at oxygen concentration 12 ppm, and then the result obtained showed that it had suffered a net weight loss rate equivalent to 6 μ/y in thickness.

This value approximately agrees with the test results of exposed non-irradiated SUS 316 specimens for 1000 hours under the same conditions in the loop. As might be estimated, it shows that neutron fluence of irradiation to this extent is not able to have any effect on corrosion. From this weight loss rate, the total released radioactivity during the test run assuming no selective leaching has taken place can be calculated. According to this estimation, approximate 0.7 mCi (0.16% in weight loss of the specimen by exposure) of radioactivity with 51Cr and 60Co as the major nuclides are assumed to transfer and to be deposited in almost entire region of the loop.

Deposition of radioactive corrosion products

(1) Corrosion products of Cr, Co, Mn, Fe in sodium are disposed to be deposited not only on low temperature region and in cold trap.
easily, but also on the wall of all tubings and components in the entire loop system. Moreover, it is not necessarily the fact that some nuclides of corrosion products are deposited in large quantity especially in the low temperature regions. (Figs. 3, 5, 6 and Photo 3) On the other hand, contents of these metallic elements in sodium is extremely small comparing with these deposited amounts. This indication was brought as the result of on-line T-ray measurement performed after having drained sodium at the initial and last stage of the experiment, showing the evidence that solubility of these corrosion products in sodium is extremely low. (Figs. 5, 6)

(2) There was found in the loop considerably defined area where particular kinds of nuclides were preferentially deposited. Cr and Co show very similar behavior of deposition, and these nuclides are predominantly deposited at the immediate downstream from the activated source specimen with little regard to temperature gradient probably by such a reaction as isotopic interchange.

There are also indicated analogous behavior between Mn and Zn. Mn is on the whole distributed throughout the loop. But more in detail, deposition behavior of this nuclide as the local figure in the loop is rather dependent upon temperature gradient (corresponding to heat exchanger): it prefer to be deposited on lower temperature region showing rapid precipitation. In isothermal region, this nuclides is easier to be deposited at the upper stream as like as Cr and Co. On the other hand, Zn is relatively readier to dissolve in sodium, and not trapped sufficiently even in the lowest temperature region like cold trap. (Figs. 3, 5, 6, 7)

(3) Generally, corrosion products preferentially are trapped and deposited in the particular parts of higher turbulent flow of the loop. (Fig. 6 and from the results of dosimeter survey throughout the loop after the operation which data are not shown in this report.)

(4) Cold trap does not deposit and remove any radioactive corrosion products very effectively from the loop. (Fig. 6)

(5) There are some regions in the loop where both corrosion and deposition occur simultaneously. In other words, radioactivity transfer does not always agree with gross mass transfer. Deposited corrosion products sticking to the surface of piping and specimens diffuse inside the wall and are hard to be removed or scaled off. Of course, such deposition cannot be washed off by liquid sodium nor by alcohol and/or water treatment.

radioactive sodium analysis

(1) There are not detected, in vacuum-distilled sodium sample taken from the expansion tank which is installed farther downstream from the radioactive source, any radioactivity of metallic impurities are derived from the major constituent elements of stainless steel.

(2) On the other part, however, traces of \(^{124}\text{Sb}, ^{113}\text{Sn}, ^{65}\text{Zn}\) and \(^{110}\text{mAg}\), solubility of which are comparatively higher were detected in the sample. (Fig. 8) These are the induced-radioactivity from the least amount of impurities in the activated specimen. The existence of Sb and Sn was confirmed as the result of another short term irradiation by JMTR of a test piece taken from the same material of which the activated specimen has been employed for this experimental run. Zn and Ag, because of these extremely small amount, have not been detected by such a method. But these nuclides are definitely not from contamination during the irradiation process of the specimen. (Figs. 9, 10, and Table I)

(3) Although sodium sampling analysis above stated results in absence or extremely small contents of Co and Mn, and also very low activity concentration of these nuclides in sodium sampling are presumed or have been realized in the primary system of LMFBR's, these nuclides
because of longer half-life could cause apprehensible concern of build-up seriously high radiation field in the primary system for longer operating period. (Figs. 9, 10)

SUBJECT TO BE INVESTIGATED IN THIS STUDY

The behavior of corrosion products in flowing sodium have been demonstrated considerably in qualitative characteristics through these experiments. Accordingly, it is necessary as a future work to undertake quantitative analysis and to understand its basic mechanism more accurately.

What have been left undetermined or not precisely analyzed in these test runs are summarized as follows:

(1) The released and deposited amounts of radioactive materials respectively, and the condition in which thermodynamic equilibrium between the release and dissolution of these materials attained in the corrosion sites of the loop.

(2) Definite factors effectively dominating corrosion and mass transfer, and explicit indication of the allowable upper level of oxygen concentration from these point of view.

(3) Accurate prediction of leaching rate of corrosion products by considering the initial or transitional period, and selectivity of the leaching nuclides. For this purpose, more detailed metallurgical analysis of the exposed specimens and further experiments of longer period are necessary.

(4) To what extent the cold trap operation does actually affect the corrosion process, or in cold trap what amounts of corrosion products can be removed from the entire system.

(5) Estimation of the effect of sodium flow velocity on the deposition.

(6) Follow-up of behavior of Ni and Fe which are, as well known, important source nuclides of induced $^{60}\text{Co}$ and $^{54}\text{Mn}$ respectively.

(7) The content of Zn which induce traceable radioactivity in stainless steel. This nuclide, even in small concentration in material, is confirmed to be easily dissolved in sodium.

CONCLUSION

The results made clear in the series of test runs of AMTL-1 are summarized as follows:

A conceptional knowledge as to deposition behavior of major radioactive corrosion products was obtained together with corrosion rate of stainless steel specimens in flowing sodium, although the test runs were carried on only for total 3000 hours. These are: the preferential deposition regions in the loop and the selectivity of the radionuclides deposited on such regions.

Using radioactivity tracer method along with on-line $\gamma$-ray spectrometry method, mass transfer phenomena in sodium system can be clarified in dynamic mode in a comparatively short period. Also, this experiment was helpful in acquiring knowledge and data relating to radiation counting method on the operating sodium loop, radioactive sodium sampling and, analytical method of radioactive sodium which are useful in the analysis for the study of radionuclide behavior in sodium system.

Principal problems concerning this research of corrosion product behavior hereafter to be solved were found out. And these are remarked as follows:

(1) Exact location or accurate range where corrosion actually occurs, and probable leaching rate in this area as well as distribution pattern of deposition of corrosion products in sodium system. (Make-up of correct model of mass transfer taking consideration into long period effect on this phenomena).
(2) Quantitative assessment of selectivity of leaching nuclides.

(3) Effect of sodium flow on deposition, also quantitatively.

(4) Formation, chemical composition, and accumulated limitations of corrosion products deposited in hot and cold regions of the loop.

(5) Origin, characteristics, transport, and content level, of particulate impurities in flowing sodium.

(6) More accurate solubility data of metallic impurities in sodium.

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Table 1 Inventory of Activated Cladding Tube

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Exposed Area (cm²)</th>
<th>Weight (g)</th>
<th>Produced by Flux of</th>
</tr>
</thead>
<tbody>
<tr>
<td>SUS316</td>
<td>6.3 × 0.35 × 10⁻⁴</td>
<td>40</td>
<td>5.34 yr</td>
</tr>
</tbody>
</table>

Sodium flux: 4 × 10¹⁰ n/cm² Th, 9 × 10¹⁰ n/cm² F > 1 Mev

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Fig. 1 Flow Diagram of Activated Material Test Loop (Mark-I)

Fig. 2 Variation of Accumulated ⁵¹Cr, ⁶⁰Co, ⁵⁴Mn on Hot leg
cold trap operated
at 200°C
flow rate 0.6 /min

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sodium drain
and recharge

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 run start
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sodium flow

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Fig. 3

Variation of Accumulated $^{51}$Cr, $^{54}$Co In Cold Trap

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Fig. 4 Deposition Profile of Radionuclides Exemplified by Exposed Stacimens

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Fig. 5 Distribution of $^{54}$Co, $^{54}$Mn Deposited on the Pipe Wall in the Main Flow Circuits

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Fig. 6 Distribution of Deposited Radionuclides along the Pipes in the Purification Circuits of AMTL-I
Fig. 7: Variation of Accumulated $^{65}$Mn, $^{65}$Zn in Cold Trap

- Sampling Na: Hot Run sampling tube No. 12
- 30 gr. at 300°C
- Spectrometry: April '72 10:00 ~ 252.5K sec
- Background radiation: 0.186

Fig. 8: $\gamma$-ray Spectrum of Vacuum distilled sodium Sample taken pulse height [channel number]
Fig. 9  Neutron-Induced γ-ray Spectrum of SUS316 Cladding Tube  (3 days after irradiation)

Sample: SUS316
Date: Jan 19, '73
Count Time: 5000 SEC
Position: 25 cm
Detector: Ge (Li)

- $6.3 \times 10^2$ x $0.35 \times 10^9$
- JRR-2
- JAN 16, '73
- 20 MIN IRRADIATION
Fig. 10  
Neutron-Induced γ-ray Spectrum of SUS316 Cladding Tube  
(10 weeks after irradiation)
Photo. 1  Cross-sectional View and Compositional Change of SUS-316 Specimen by Scanning Electron Microprobe with non-Dispersive Si (Li) X-ray Detector  
—Exposed to 650°C, 2.5m/sec Sodium for 1000hr—

Photo. 2  Scanning Electron Microprobe Analysis of Depleted Surface of SUS-316 Specimen  
—Exposed to 650°C, 2.5m/sec Sodium for 1000hr—

Photo. 3  Scanning Electron Microprobe Analysis of Corrosion Product Deposited on SUS-316 Specimen  
—Exposed to 500°C, 0.9m/sec Sodium for 1000hr—