Cold Trapping of Fission Products in a Stainless Steel Sodium Loop

Norimasa MITSUTSUKA, Hikaru SHIMOJIMA, Yohichi GOHSHI,
Tokyo Shibaura Electric Co., Ltd.*
Horst FEUERSTEIN
Gesellschaft für Kernforschung m.b.H.**

Received May 1, 1976
Revised September 10, 1976

The behavior of fission products in sodium system was studied using a cold trap installed in the Toshiba Fission Product Loop, which is a stainless steel inpile sodium loop. The fission products were generated within the liquid sodium by irradiating UO₂ fuel under sodium convection. The cold trapping behavior of ⁹²Zr, ⁹⁵Nb, ¹³²Te, ¹³¹I, ¹³⁷Cs and ¹⁴⁰La (¹⁴⁰Ba) was examined by measuring the γ-rays in the cold trap and in the sodium dump tank by means of Ge(Li) detectors. Such fission product nuclides as ¹³¹I, ¹³²Te and ¹³⁷Cs were found to be reversively removed by the cold trap at lower temperatures, with negligible hysteresis. The distribution coefficient $K(cm)$ for the cold trap surface was found to be expressible by $\log K = (3.08 \pm 0.17) - (0.0112 \pm 0.0006) \times T(°C)$ for ¹³¹I, and by $\log K = -(4.55 \pm 0.29) + (1830 \pm 140) \times 1/T(K)$ for ¹³⁷Cs. The behavior of ¹³¹I and ¹³⁷Cs during cold trapping can be explained clearly by adsorption model using these $K$ values. While ⁹²Zr, ⁹⁵Nb, ¹⁰³Ru and ¹⁴⁰La (¹⁴⁰Ba) also were detected in the cold trap, the efficacy of cold trapping is far lower for these nuclides than for ¹³¹I and ¹³⁷Cs, and changing the cold trap temperature did not appear to have any appreciable effect on the cold trapping. Presence of ¹⁴¹Ce and ¹⁴⁴Ce was not detectable in the cold trap, although small amounts of these nuclides were detected in the sodium dump tank.

KEYWORDS: fission products, liquid metals, sodium, inpile sodium loop, cold traps, distribution coefficient, iodine 131, cesium 137, tellurium 132

I. INTRODUCTION

The behavior of fission products (F.P.) in stainless steel-sodium systems is particularly important in radioactive contamination studies of LMFBR (Liquid-Metal-cooled Fast Breeder Reactor) primary systems. With the view to obtaining data on this aspect, a stainless steel inpile sodium loop—FPL (Fission Product Loop)—has been installed on the Toshiba Training Reactor (TTR-1)(¹). The present paper describes the results obtained from F.P. cold trapping experiments performed using the FPL cold trap.

Cold traps are used in LMFBR’s for the purpose of sodium purification, mainly to remove oxygen, but also for carbon and hydrogen elimination. Cold traps are also known to be effective in removing certain F.P. from sodium. Cold trap operating experience in sodium or NaK cooled reactors has been reported in literature.(²) A number of capsule and loop experiments are also known to have been conducted on certain F.P. with use made of radioactive tracers.(³) These studies, however, have not yet come to clarify the processes and mechanisms of cold trapping except in respect of certain F.P. While several reports have been published on reactor operating experience data

* Ukishima-cho, Kawasaki-ku, Kawasaki-shi.
** Karlsruhe, GERMANY.
associated with F.P. behavior, they are not systematic, and generally inadequate for detailed analysis of cold trapping behavior. Tracer experiments may be useful for some but not for all the nuclides requiring consideration in the LMFBR: Difficulties arise when attempting to prepare radioactive tracers in the same chemical and carrier-free form as found in the LMFBR.

Reactor irradiation of UO₂ fuel in the FPL under sodium convection is found to release F.P. into the sodium, the mechanism being probably recoil. This suggests that most F.P., to be considered in LMFBR's, could be prepared by this means in the sodium itself in completely carrier-free form, and under conditions much more closer to those prevailing in LMFBR's. The present work concerns a study using the FPL cold trap loop to examine the cold trapping behavior of F.P. in sodium, prepared by irradiating the FPL fuel.

II. EXPERIMENTAL

1. FPL Cold Trap Loop

The present experiments were performed using the Toshiba Fission Product Loop (FPL), details of which have been reported elsewhere. The FPL consists of two independent stainless steel sodium loops.—(a) for inpile experiments, containing natural UO₂ fuel, (b) for sodium purification experiments with cold trap. These two loops are connected to the sodium dump tank. Fission products are prepared by irradiation applied to the inpile loop under sodium convection.

Figure 1 represents the flow diagram of the FPL cold trap system. The cold trap (CT) is of conventional cylindrical form and contains a stainless steel wire mesh of 0.35 g/cm³ density. The trap is cooled by air blower, the trap temperatures being measured at two positions with thermocouples (TC) inserted into wells of different depths. The dump tank (DT) and loop piping temperatures were also monitored by thermocouples suitably arranged. An electromagnetic pump (EMP) recirculates the sodium containing the F.P. through the cold trap via a regenerative heat exchanger (HX). The sodium flow rate is measured by electromagnetic flowmeter (EMF).

Table 1 summarizes some of the relevant parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sodium inventory</td>
<td>4.8 kg</td>
</tr>
<tr>
<td>Impurity level</td>
<td>~80 ppm by weight (oxygen)</td>
</tr>
<tr>
<td>Cover gas</td>
<td>Argon</td>
</tr>
<tr>
<td>Main structural material</td>
<td>AISI 304</td>
</tr>
<tr>
<td>Main pipe</td>
<td>13.8 mm O.D., 2.2 mm thick</td>
</tr>
<tr>
<td>Cold trap volume</td>
<td>930 cm³</td>
</tr>
<tr>
<td>Cold trap surface</td>
<td>6,000 cm²</td>
</tr>
<tr>
<td>Cold trap packing</td>
<td>Stainless steel wire mesh</td>
</tr>
<tr>
<td>Sodium flow rate</td>
<td>≤5 l/min</td>
</tr>
<tr>
<td>Temperature</td>
<td>≤500°C</td>
</tr>
<tr>
<td>UO₂ inventory (inpile loop)</td>
<td>150 g in natural uranium content</td>
</tr>
<tr>
<td>Neutron flux (inpile loop)</td>
<td>~10⁶ n/cm²·sec</td>
</tr>
</tbody>
</table>

2. Fission Products Preparation

Prior to commencing the cold trapping experiments, the inpile loop was irradiated...
for 4 hr at full reactor power (100 kW) under sodium convection. A fraction of the F.P. produced in the UO₂ fuel was released directly into the liquid sodium by recoil release. The irradiated sodium was usually drained into the sodium dump tank within 6 to 12 hr after the end of irradiation. The sodium was left to stand in the dump tank for 6 days at room temperature in order to reduce gross Na radioactivity, upon which its activity decreased to the same order of intensity as that of the F.P. This left the dump tank sodium loaded with completely carrier-free F.P., ready for the cold trapping experiments.

3. Cold Trap Operation
A cold trapping experiment was usually begun by flushing the cold trap with hot sodium for more than 4 hr at a temperature near 400°C. Upon this flushing, the oxygen concentration of the sodium was found to be about 80 ppm by weight, as determined by a plugging indicator attached to the inpile loop. The cold trap temperature was changed almost stepwisely by changing the air blower speed. Temperature equilibrium was usually established within 1 hr, after which the cold trap temperature was maintained constant for more than 3 hr. Dump tank heating was continued throughout a run, but its temperature was changed along with the cold trap temperature. At a cold trap temperature of 120°C, the dump tank temperature lowered to 280°C at a sodium flow rate of 0.7 l/min and to 220°C at 1.5 l/min. Figure 2 shows a typical record of the operating temperatures at various points along the loop under a sodium flow rate of 0.7 l/min.

4. Radioactivity Measurements
The γ-ray spectra were measured at the dump tank and at the cold trap, using Ge(Li) detectors and a 4096 channel pulse height analyzer with magnetic tape output capability. Gamma ray spectrum data obtained during the cold trapping experiments were stored in magnetic tape, which was subsequently used for offline computer processing.

III. RESULTS
1. Fission Products detected in Dump Tank and in Cold Trap
After cooling in the dump tank for 6 days, as already mentioned, measurements were made of the γ-ray spectra of the F.P. contained in the sodium. The relative intensities of radioactivity of the F.P. entrained in the sodium differed from the values calculated on the basis of the relevant irradiation and cooling conditions: ¹³¹I and ¹³²Te/¹³²I showed the strongest radioactivity. The radioactivity of ¹⁰⁶La(¹⁰⁶Ba) was lower than those of ¹³¹I and ¹³²Te/¹³²I by almost one order of magnitude, while other nuclides, which were expected to show still higher radioactivity, such as ⁹⁹Mo, ¹⁴⁷Ce and ¹⁴⁷Nd, were not even detected. On the other hand, some long-lived nuclides were detected, which were considered to have been accumulated during previous irradiations: Compared with ¹³⁷Cs, the radioactivities of ⁹⁵Zr, ⁹⁵Nb, ¹⁰⁹Ru, ¹⁴¹Ce and ¹¹⁴Ce were lower with a difference larger than one order of magnitude.

The differences between the calculated and observed relative radioactivities are considered to derive from the affinities of the F.P. with liquid sodium. Iodine and tellurium are known to dissolve in liquid sodium to form sodium compounds. Cesium mixes well with liquid sodium. The other F.P., which showed lower relative radioactivities, are, generally speaking, those that show very low solubility in sodium: The inpile loop piping after long cooling time showed a strong deposition of F.P. such as ⁹⁵Zr, ⁹⁵Nb, ¹⁴¹Ce and ¹¹⁴Ce. Thus the residual radioactivity of the F.P. that showed lower values of radioactivity in the drained sodium, was considered to
have deposited around the inpile loop piping due to poor affinity with liquid sodium or through strong adsorption on the inner surface of the pipes.

Some F.P. had already deposited in the cold trap during previous sodium cleanup operations. The dominantly present nuclide detected in the cold trap was $^{131}$I. A small amount of $^{95}$Zr, $^{95}$Nb, $^{103}$Ru and $^{140}$La($^{146}$Ba) were also detected, while $^{141}$Ce and $^{144}$Ce were not discernible. Upon flushing the cold trap with hot sodium, a considerable amount of $^{131}$I, $^{133}$Te/$^{132}$I and $^{137}$Cs redissolved into the sodium and returned to the dump tank, while the trapped $^{95}$Zr, $^{95}$Nb, $^{103}$Ru and $^{140}$La($^{146}$Ba) mostly remained in the cold trap, even after flushing with sodium at 400°C.

2. Fission Product Cold Trapping Behavior in Sodium

Figure 3 shows the typical cold trapping behavior of $^{131}$I at a sodium flow rate of 1.5 l/min, and evidences the very effective removal of this nuclide by cold trapping. A step reduction in trap temperature is seen to have been followed by a fairly quick response of $^{131}$I radioactivity drop, and equilibrium would appear to have been established within 3 hr after each temperature change. About 30% of the $^{132}$Te in the dump tank was removed at a cold trap temperature of 280°C,

![Fig. 3 Cold trapping behavior of $^{131}$I](image)

Fig. 4 Fission product radioactivity at different cold trap temperatures
but further lowering below this level did not enhance $^{132}$Te elimination to any appreciable extent. The $^{137}$Cs in the dump tank did not change appreciably at higher cold trap temperatures, but at 120°C, about 70% of $^{137}$Cs in the dump tank was gradually removed. The other nuclides, $^{95}$Zr, $^{95}$Nb and $^{140}$La(140Ba) did not appear to show any appreciable response to cold trapping operation, either at the cold trap or at the dump tank. The data on $^{95}$Zr, $^{95}$Nb and $^{140}$La(140Ba) scattered prominently, due to the low counting rates, but in so far as discernible, the cold trap appeared ineffective for the removal of these nuclides from the sodium in the dump tank. In order to examine the reversibility of the cold trap, the tests were extended into a reheating course as depicted in Fig. 2. Figures 4(a) and (b) present the F.P. radioactivities measured in the dump tank and in the cold trap at different trap temperatures. The open marks represent the average count rates obtained during cooling, and the closed marks the corresponding data for the reheating course. It is seen that the reheating plots retrace the downward steps quite faithfully in the cases of $^{131}$I, $^{132}$Te and $^{137}$Cs.

3. Sodium Draining Tests

The radioactivity measured in the cold trap under sodium convection would contain contributions from both trapped and dissolved F.P. In order to distinguish between these two kinds of F.P., additional tests were undertaken, in which, with all other conditions maintained unaltered, the cold trap sodium was drained after being left standing for about 5 hr at the relevant cold trap temperature under sodium convection, and the $\gamma$-ray spectra were measured at both cold trap and dump tank during the sodium convection and after sodium draining. The results are presented in Figs. 5(a) and (b) in the same form as the two preceding figures, but for the

![Fig. 5 Fission products radioactivity before and after sodium draining](image-url)
cooling course only, and separately for the periods during sodium convection and after sodium draining from the trap. After draining, the sodium was allowed to stand for about 20 min, in order to achieve radioactive equilibrium between $^{137}$Cs and $^{137m}$Ba.

The plots of Fig. 5(a) indicate that more than 80% of the $^{24}$Na was entrained out of the cold trap with the draining. Similarly, about 80% of $^{137}$Cs and 50% of $^{131}$I and $^{132}$Te were entrained out when the trap temperature was 390°C, but the fraction entrained of these three nuclides diminished with decreasing trap temperature, and below 280°C, the count rates came to be higher after than before draining, in the cases of $^{131}$I and $^{132}$Te. This inversion can be ascribed to removal of the shielding effect that had been provided by the sodium while present in the trap, while the fractions of these nuclides entrained out with the draining were negligibly small.

The remaining nuclides $^{90}$Zr, $^{92}$Nb and $^{140}$La ($^{140}$Ba) showed little propensity to be entrained with the sodium except at the highest trap temperatures, at which an appreciable amount of entrainment is indicated from both Figs. 5(a) and (b). The vertical bars drawn on the plots reveal that the plots scattered considerably on account of the poor counting rates. These nuclides did not manifest any marked change during sodium convection. They must have been affected in some way during the draining of the sodium from the cold trap at the higher temperatures.

4. Cold Trapping Behavior of $^{103}$Ru, $^{141}$Ce and $^{144}$Ce

The radioactivities of $^{103}$Ru, $^{141}$Ce and $^{144}$Ce were too low to be discerned above the gross $^{24}$Na activity after the 6 days cooling, so additional runs were conducted with 30 days cooling time, in order to obtain some information on the cold trapping behavior of these nuclides. The radioactivities observed in the loop during the period that followed this cooling are shown in Fig. 6 for $^{103}$Ru, $^{141}$Ce, $^{144}$Ce, $^{95}$Nb and $^{137}$Cs, together with the levels measured in the loop before sodium charging and after draining off. The corresponding cold trap temperatures are shown at the bottom of the figure. The trap temperature was changed gradually from 300°C to 110°C and was then kept at 110°C for 6 hr. No marked change is seen throughout the period covered in the plots of $^{103}$Ru, $^{141}$Ce and $^{144}$Ce as it is the case also of $^{95}$Nb, whereas $^{137}$Cs has undergone a significant decrease with lowering cold trap temperature.

![Fig. 6 Cold trapping behavior of long-life F.P.](image)

5. Cold Trapping Behavior of $^{137}$Cs

Cesium-137 is in radioactive equilibrium with its decay product $^{137m}$Ba and is measured by counting the $^{137m}$Ba, whose half-life is only 2.6 min, calling for special care in the measurement under flowing sodium. The effects of sodium flow rate on the cold trapping behavior of $^{137}$Cs/$^{137m}$Ba were examined at the end of a long (6 months) reactor shutdown. Such an occasion favored the determination of $^{137}$Cs/$^{137m}$Ba, since it then constituted the major radioactivity in the loop, and the reduced interference of other nuclides produced better counting statistics.

Figure 7 shows the effect of changes in the sodium flow rate on the $^{137m}$Ba count rate in the dump tank. Loop temperatures also changed with the sodium flow rate, but the cold trap temperature was kept constant within ±5°C. It is seen that the $^{137m}$Ba count
rate was strongly influenced by the sodium flow rate, with a distinct decrease with increasing flow rate at the higher trap temperatures. This behavior can be ascribed to the transport of $^{137m}$Ba to the cold trap along with the liquid sodium, and with its strong propensity to deposit, the $^{137m}$Ba thus transferred into the cold trap collected on the trap surface, even at high trap temperatures.

Figure 8 shows the $^{137}$Cs/$^{137m}$Ba count rate in the dump tank, measured after letting the sodium stand for about 30 min after stopping the sodium flow to allow radioactive equilibrium to be attained between $^{137}$Cs and $^{137m}$Ba. The count rate changed with cold trap temperature in roughly the same manner as with 0.7 l/min sodium convection in the earlier reversibility tests. This would imply that the cold trapping behavior of $^{137}$Cs at a sodium flow rate of 0.7 l/min, as determined by $^{137m}$Ba counting, could be validly utilized as measure for knowing the $^{137}$Cs behavior.

### IV. DISCUSSION

#### 1. General Considerations

Table 2 summarizes the ratios obtained for the F.P. count rates between that in the cold trap and that in the dump tank. The count rates at these two positions being roughly equal for $^{24}$Na, these ratios would also serve as indication of the relative efficacy of the cold trap for removing the relevant F.P., at the indicated trap temperatures. The data given in this table concur with those from other cold trapping experiments in indicating the existence of two distinct groups of F.P. in the FPL cold trap loop: (a) Those susceptible to reversible cold trapping—such as $^{131}$I, $^{133}$Te and $^{137}$Cs, and (b) those not susceptible to cold trapping—such as $^{69}$Zr, $^{95}$Nb, $^{103}$Ru, $^{140}$La($^{140}$Ba), $^{141}$Ce and $^{144}$Ce. Major fraction of the F.P. of the second group remains within the inpile part of the FPL and

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Radioactivity ratio (CT/DT)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{131}$I</td>
<td>0.8</td>
</tr>
<tr>
<td>$^{133}$Te</td>
<td>0.9</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>0.7</td>
</tr>
<tr>
<td>$^{69}$Zr</td>
<td>0.5</td>
</tr>
<tr>
<td>$^{95}$Nb</td>
<td>1.1</td>
</tr>
<tr>
<td>$^{140}$La($^{140}$Ba)</td>
<td>0.7</td>
</tr>
<tr>
<td>$^{103}$Ru</td>
<td>— *&lt;sup&gt;††&lt;/sup&gt;</td>
</tr>
<tr>
<td>$^{141}$Ce</td>
<td>— *&lt;sup&gt;††&lt;/sup&gt;</td>
</tr>
<tr>
<td>$^{144}$Ce</td>
<td>— *&lt;sup&gt;††&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

* After reheating cold trap
*<sup>††</sup> Not detected due to interference with $^{24}$Na
*<sup>***</sup> Detected in dump tank, not detected in cold trap
only a small portion is transferred to the cold trap.

The foregoing results are generally consistent with those reported from the operating experience of cold traps in sodium or NaK cooled reactors or from capsule experiments\(^2\)~\(^9\). The results are discussed in more detail in the following sections.

2. Distribution Coefficients of F.P. on Cold Trap Surface

Cold traps in sodium systems usually remove impurities from liquid sodium by precipitation, as typically represented in oxygen removal. Under the present experimental conditions, in which F.P. were released by direct recoil from UO\(_2\) fuel into liquid sodium, their concentrations in liquid sodium were far lower than their solubilities. Hence precipitation in the cold trap could not occur even at the lowest cold trap temperature. In such a case, adsorption on the cold trap surface comes to play an important role. Thus the removal of \(^{131}\)I, \(^{133}\)Te and \(^{137}\)Cs by the cold trap is considered to be due to adsorption on the stainless steel surfaces of the cold trap. Such adsorption on the trap surfaces is expressed in terms of a distribution coefficient defined by

\[
K = \frac{\text{F.P. weight at surface}}{\text{F.P. weight in sodium}} \cdot \frac{\text{Unit area}}{\text{Unit volume}}. \tag{1}
\]

It being difficult to determine \(K\) directly from the above expression, it is derived from the results of sodium draining tests, through the formula

\[
K = \frac{\text{Radioactivity remaining on CT surface (cps)}}{\text{Radioactivity drained with sodium (cps)}} \cdot \frac{1}{\text{Cold trap surface to volume ratio (cm}^{-1})}. \tag{2}
\]

For \(^{131}\)I, \(^{137}\)Cs and \(^{133}\)Te, \(K\) is calculated from the change in radioactivity in the cold trap and in the dump tank, observed at the time of sodium draining.

(1) \(K\) value of \(^{131}\)I

The \(^{131}\)I radioactivity observed in the cold trap was lower during sodium convection than after sodium draining at lower cold trap temperatures, due probably to shielding by the liquid sodium in the cold trap. However, significant increase of \(^{131}\)I radioactivity was observed in the dump tank after draining the sodium in the cold trap. This increase was used for calculating the \(^{131}\)I radioactivity drained with the sodium. The radioactivity remaining dissolved in the sodium contained in the cold trap was corrected also based on the remaining \(^{24}\)Na radioactivity. Figure 9 presents the values of \(K\) for \(^{131}\)I thus derived, plotted against the reciprocal of trap temperature. The straight line drawn through these plots represents the least square fitting expressed by

\[
\log K (\text{cm}) = -(5.61 \pm 0.32) + (2.95 \pm 0.15) \times 10^3 \cdot 1/T(K). \tag{3}
\]

The dashed line in the figure shows the \(K\) function by Allan\(^3\), which represents \(K\) against temperature expressed in centigrade. The present data also fit better against centigrade temperature:

\[
\log K (\text{cm}) = -(3.08 \pm 0.17) - (0.0112 \pm 0.0006) \cdot T(\text{°C}). \tag{4}
\]

This fit is shown in Fig. 9 by the solid curve, and reveals good consistency with Allan's \(K\) function.
Allan derived his function from experiments with high specific radioactivity tracer, which can be considered to yield much the same results on iodine behavior in liquid sodium as obtained using iodine released by the fuel in the sodium loop. A difference between Allan's work and the present, in terms of experimental conditions, is the oxygen level, which was below 20 ppm for Allan's and was 80 ppm for the present study (both by weight), but this extent of difference would not appear to influence the behavior of iodine to any appreciable degree.

(2) K value of $^{137}\text{Cs}$

As already mentioned, the $^{137}\text{Cs}$ radioactivity was determined by measuring its decay product $^{137m}\text{Ba}$, and it was noted in Sec. III-5 that the count rate of $^{137m}\text{Ba}$ under sodium convection was strongly affected by the sodium flow rate. Hence, the count rates of $^{137m}\text{Ba}$ after sodium draining were only valid for the determination of $^{137}\text{Cs}$ at the cold trap surface. The radioactivity of $^{137}\text{Cs}$ dissolved in the sodium was calculated using the radioactivity of $^{137}\text{Cs}$ at the dump tank after sodium draining, assuming that all the $^{137}\text{Cs}$ detected at the dump tank had dissolved in the sodium. This assumption can be considered valid by the fact that the dump tank temperature was kept above 260°C during the sodium draining tests, under which condition most of the $^{137}\text{Cs}$ in the dump tank can be expected to have dissolved in the sodium, with negligible deposition on the inner surface of the dump tank.

Results are shown in Fig. 10 in presentation similar to the preceding figure. Least square fitting yielded the formula

$$\log K(\text{cm}) = -(4.55 \pm 0.29)$$
$$+ (1830 \pm 140) \cdot 1/T(K). \quad (5)$$

The importance for LMFBR's of $^{137}\text{Cs}$ behavior in sodium systems has motivated intensive studies in this domain$^{(6)}$-$^{(8)}$, which have resulted in several $K$ values reported for cesium in stainless steel-sodium systems. The $K$ function reported by Cooper & Taylor in the figure is that for as received stainless steel at a cesium concentration of 0.14 ppm (atom) in sodium$^{(7)}$. Their function shows excellent consistency with the present $K$ function, which latter can thus be considered to be valid, at least up to 0.14 ppm (atom) of cesium in liquid sodium. In the present instance, $^{137}\text{Cs}$ must have been in concentrations below $10^{-3}$ ppm (atom).

![Fig. 10 Distribution coefficient for $^{137}\text{Cs}$](image)

Reports have mentioned the occurrence of surface deactivation after exposure to high temperature sodium (1,000°F)$^{(6)}$-$^{(8)}$. The single- and double-dotted chain lines in Fig. 10 represent Guon's $K$ functions for activated and deactivated surfaces, respectively$^{(9)}$. In the present instance, no such surface deactivation was observed even after 2 years of experiment. The cold trap was never heated beyond 420°C, so that heat cycles conducted below this temperature would not appear to have any influence on the adsorption of cesium on the stainless steel surface.

(3) $K$ value of $^{132}\text{Te}$

The $K$ value was calculated for $^{132}\text{Te}$ in the same manner as for $^{131}\text{I}$, with the result shown in Fig. 11. The plots, apart from scattering appreciably, do not appear to suggest the existence of a straight line between $\log K$ and $1/T(K)$. And in fact no report is found in literature giving the value of $K$ for tellurium in a stainless steel-sodium system. The only mention in this connection is that of a high tendency of $^{132}\text{Te}$ to deposit
in a NaK system\(^{(9)}\). In the present case, \(^{132}\)Te showed strong adsorption on the cold trap surface at higher temperatures, while below 250°C the adsorption became constant. No plausible explanation can be offered for this behavior at the present time. Oxygen impurities in the cold trap loop begin to precipitate at a cold trap temperature of 280°C. This makes it necessary to examine the oxygen precipitation effect on tellurium adsorption on a stainless steel surface.

No plausible explanation can be offered for this behavior at the present time. Oxygen impurities in the cold trap loop begin to precipitate at a cold trap temperature of 280°C. This makes it necessary to examine the oxygen precipitation effect on tellurium adsorption on a stainless steel surface.

3. Application of Adsorption Model to Cold Trapping Behavior Analysis of \(^{131}\)I and \(^{137}\)Cs

The observed deposition of \(^{131}\)I and \(^{137}\)Cs at the cold trap was confirmed to be due to adsorption on the cold trap surface. Calculations based on an adsorption model and using derived values of \(K\) can be applied to cold trapping behavior analysis of \(^{131}\)I and \(^{137}\)Cs.

After establishment of steady forced convection of liquid sodium, the concentration \(c_i\) of the F.P. \(i\) in the sodium can be assumed to be constant for all parts of the loop. The amount \(a_{ij}\) of the F.P. \(i\) adsorbed in the region \(j\) of the loop is derived from the \(K\) function, and knowing also the surface area \(S_j\) and the temperature \(T_j\) of the same region \(j\):

\[
a_{ij} = K_j(T_j)S_j c_i, \quad (6)
\]

The total amount \(A_i\) of adsorbed F.P. can be calculated by integrating the equation around the loop:

\[
A_i = \sum_j K_j(T_j)S_j c_i, \quad (7)
\]

and the total dissolved F.P.

\[
C_i = c_i V, \quad (8)
\]

where \(V\) is the total sodium volume. The total inventory \(I_i\) of the F.P. \(i\) can be assumed to be constant during the cold trapping:

\[
I_i = A_i + C_i = \text{const.} \quad (9)
\]

The fractions \(C_i/I_i\) of \(^{131}\)I and \(^{137}\)Cs dissolved were determined using the derived values of \(K\), from calculations for which the cold trap loop was divided into 10 regions. The solid lines in Fig. 12 represent the calculated \(^{131}\)I and \(^{137}\)Cs fractions vs. reciprocal of cold trap temperature. The count rates of the \(^{131}\)I and \(^{137}\)Cs observed in the dump tank during the cold trapping, are superimposed in the figure. It is seen that the dump tank radioactivity represents quite well the dissolved radioactivity. The \(^{131}\)I radioactivities observed at lower temperatures are not reproduced in the figure, because an appreciable amount of \(^{131}\)I becomes adsorbed on the inner surface of the dump tank. The \(^{137}\)Cs radioactivity values are those observed with the sodium flow stopped.
Figure 12 indicates that the observed count rates of $^{131}$I and $^{137}$Cs in the dump tank are consistent with the calculated dissolved fractions, within experimental error. This evidences the validity of the adsorption model to explain the cold trapping behavior of $^{131}$I and $^{137}$Cs.

4. Cold Trapping Behavior of $^{95}$Zr, $^{98}$Nb, $^{103}$Ru and $^{140}$La($^{140}$Ba)

The major fraction of these nuclides released into the flowing sodium from the fuel remained within the inpile part of the FPL, deposited on the piping surface, and only a small fraction was transferred to the dump tank and the cold trap loop. These nuclides appear to have a strong tendency to adsorb on the stainless steel surface and to have very low solubility in liquid sodium. Only a small portion of these nuclides found their way into the cold trap, and the distribution ratios of these F.P. between the cold trap and the dump tank were lower than those of $^{131}$I, $^{132}$Te and $^{137}$Cs, as is seen from Table 2. The small tendency of these nuclides to pass into the cold trap is also seen to have resulted in fairly constant amounts being found in the cold trap, irrespective of trap temperature.

A slight difference in their counting rates was observed upon draining the cold trap sodium at temperatures higher than 300°C. The cause of this difference is not clear. The effect of oxygen, however, might have some relation. The cold trap loop contained about 80 ppm (weight) oxygen in the present instance, and can be considered to have dissolved in the loop sodium at sodium temperatures above 280°C. Further detailed considerations are rendered difficult by the poor counting rate, and we will limit ourselves in the present instance to the remark that the effect of oxygen dissolved in liquid sodium should not be ignored.

While the behavior of these nuclides is very important on account of their close relation to problems of radioactive contamination of LMFBR primary systems, the insufficiency of data available from the present experiments do not permit meaningful discussion of the behavior of these F.P. quantitatively. The work has nevertheless indicated that, the conventional tracer technique, which would appear quite inadequate for the preparation of these F.P. in stainless steel-sodium systems for the study of their behavior in sodium loops, might quite usefully be replaced by the use of the FPL inpile loop. An experimental program using the inpile loop of the FPL with improved counting statistics is now under way at Toshiba.

5. Cold Trapping Behavior of Cerium

Only limited experimental data were available on the cold trapping behavior of $^{141}$Ce and $^{144}$Ce, due to the low $\gamma$-ray energy and low count rate, but the information gathered still served to show that only negligible amounts of $^{141}$Ce and $^{144}$Ce were transported to the cold trap. Cerium solubility in oxygen-containing sodium systems has been reported to be low at higher sodium temperatures—a tendency contrary to what is observed in the temperature characteristics of the solubility of other nuclides in liquid sodium.

Such an element could not possibly be eliminated by cold trapping by precipitation. Cerium is also known to deposit on stainless steel surfaces in a system containing sodium. It will be recalled that in the present experiment, irradiated sodium was cooled and then left to stand at room temperature for 6 days before cold trapping. During the solidification of sodium, cerium would have deposited on the inner surface of the dump tank. This will have precluded all possibility for the cerium to be transported to the cold trap.

V. CONCLUSION

Cold trapping of carrier-free F.P. in liquid sodium was investigated, using the cold trap loop of the Toshiba FPL. The following conclusions are derived.

(1) Most of the $^{131}$I, $^{135}$Te and $^{137}$Cs was carried into the cold trap loop upon draining the sodium in the inpile loop: these nuclides were reversively removed from the liquid sodium at low cold trap temperatures with negligible hysteresis.
(2) The cold trapping mechanism of $^{131}$I and $^{137}$Cs is considered to be adsorption on the cold trap surface. The distribution function on the cold trap surface was determined for $^{131}$I and $^{137}$Cs:

$$\begin{align*}
\text{I$: } & \log K = (3.08 \pm 0.17) \\
& - (0.0112 \pm 0.0006) \cdot T(\text{°C}) \\
\text{Cs$: } & \log K = -(4.55 \pm 0.29) \\
& + (1830 \pm 140) \cdot 1/T(\text{K}).
\end{align*}$$

(3) Other nuclides mostly remained within the inpile loop as pipe deposits and only a small part was transferred to the cold trap loop: Small amounts of $^{90}$Zr, $^{92}$Nb, $^{103}$Ru and $^{148}$La($^{194}$Ba) were found to be removed by the cold trap, but the efficacy of cold trapping is far lower for these nuclides as it is for $^{131}$I and $^{137}$Cs. No traces of $^{141}$Ce and $^{144}$Ce were found in the cold trap, although small amounts of these nuclides were detected in the sodium dump tank.

ACKNOWLEDGMENT

The authors gratefully acknowledge the collaboration of Messrs. Y. Ookoshi, I. Sakaijiri, K. Ono, K. Fukushima and S. Hara of Toshiba R & D Center. Particular acknowledgment is due to Mr. H. Yamaguchi for his assistance with loop operation, and to the operating staff of TTR-1 for their collaboration with inpile irradiation.

The authors appreciate the encouragement accorded by Prof. T. Shiokawa of Tohoku University and Dr. I. Fujii of the Toshiba R & D Center.

(Text edited grammatically by Mr. M. Yoshida.)

REFERENCES