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DEPOSITION AND CONTROL OF ^{7}Be IN LIQUID LITHIUM

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DEPOSITION AND CONTROL OF ^{7}Be IN LIQUID LITHIUM

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ABSTRACT

Preferential beryllium-7 deposition has been found in the higher temperature region of non-isothermal flowing lithium between 270°C to 200°C. Various methods for controlling ^{7}Be distribution for application to the Fusion Materials Irradiation Test (FMIT) facility are examined. Flushing a loop with 425°C lithium decreased ^{7}Be activity on pipe walls by 60-80%. Yttrium has been found to be more effective than the other materials tested for removal of ^{7}Be from lithium. Preliminary results on diffusion traps indicate effectiveness for ^{7}Be removal also.

INTRODUCTION

Beryllium-7, which has a half-life of 53 days and emits a 0.48 MeV photon, will be a large source of radioactivity in the lithium system of the Fusion Materials Irradiation Test facility (FMIT).[1] In this facility the nuclide will be produced by the interaction of a 35 MeV deuteron beam with the lithium target via two reactions: $^{7}Li(d,2n)^{7}Be$ and $^{6}Li(d,n)^{7}Be$. The ^{7}Be equilibrium concentration in the FMIT facility was recently estimated at 4.54×10^{4} curies. With an active lithiume volume of $\sim$ 4500 liters, this concentration is equivalent to 10 mCi/cm^{3} of lithium. Calculations[2] show that the radiation fields near FMIT piping and components without ^{7}Be control will be 10 to 30 R/hr or higher at a distance of one foot. Radiation fields of this magnitude limit access for hands-on maintenance.

Previous work[3] identified both preferential segregation of ^{7}Be at interfaces and ^{7}Be penetration into stainless steel at 270°C over extended periods of time (4000 hours). This report describes the transport of ^{7}Be in lithium as a function of temperature. Also described is the development of several removal methods to minimize the impact of ^{7}Be transport on FMIT operations.

SUMMARY

The work reported herein was conducted in small lithium loops operating over short periods of time (1000 hrs) at 270°C in the hot leg and 200°C cold leg. It showed that ^{7}Be was deposited throughout the system piping, with more deposition occurring at higher temperatures. Methods of controlling ^{7}Be were investigated. Yttrium was the most effective deposition substrate material at 270°C for ^{7}Be removal, being more effective than pure iron, Fe-2-1/4 Cr-1 Mo steel, 304 stainless steel (SS), cobalt, nickel, and metallic beryllium. Incidentally, nickel showed severe corrosion in lithium even at 270°C.

A hot flush with lithium at 425°C removed 60-80% of the ^{7}Be deposited in loops operated for approximately 1000 hours. Since ^{7}Be diffuses into stainless steel at 270°C, the hot flush would be expected to be less effective after longer operating times.

A diffusion cold trap was effective at removing ^{7}Be from lithium, with extensive ^{7}Be deposition at the solid-liquid interface in the trap.

EXPERIMENTAL PROCEDURES

Materials

The ^{7}Be for these experiments was prepared by Oak Ridge National Laboratory (ORNL) as an unprocessed cyclotron target: $^{6}Li_3(p,\gamma)^{7}Be_4$ and $^{7}Li_3(p,n)^{7}Be_4$. Irradiated lithium was removed from a Cu backing plate and placed inside 0.2-mm mesh 304 SS bags for use in the tests described here.

Thin sheet specimens of iron, cobalt, nickel, beryllium, and yttrium(all 99+% pure material), as well as Fe-2-1/4 Cr-1Mo ferritic steel and 304 SS, were used as deposition test materials. These materials were cut into 10 mm x 24 mm specimens, of thicknesses ranging from 127 μm to 635 μm. All samples in the initial test were washed in Na_3PO_4 solution, rinsed in de-ionized water, and dried in vacuo before weighing. All ferrous specimens in the second test were electropolished (16V) with 10% perchloric acid in glacial acetic acid, and the Be was electropolished with 10% perchloric acid in butanol. All yttrium specimens were cleaned in 10% KOH solution.

Test Loops and Operations

As illustrated in the schematic in Figure 1, two small loops were constructed from type 304 H stainless steel tubing of the following dimensions: 3/4-in. O.D., 35-mil wall (19-mm O.D., 0.9-mm wall). A linear induction motor was used to pump the lithium. Cooling fins were tack-welded to the cold leg test section to assist with heat dissipation. The two vertical sections, each 38-cm long, contained thermowells for monitoring temperature along the test sections. Deposition coupons were attached to each thermowell with stainless steel pins tack-welded to the tube. The total volume of each loop was approximately 0.4 liter.

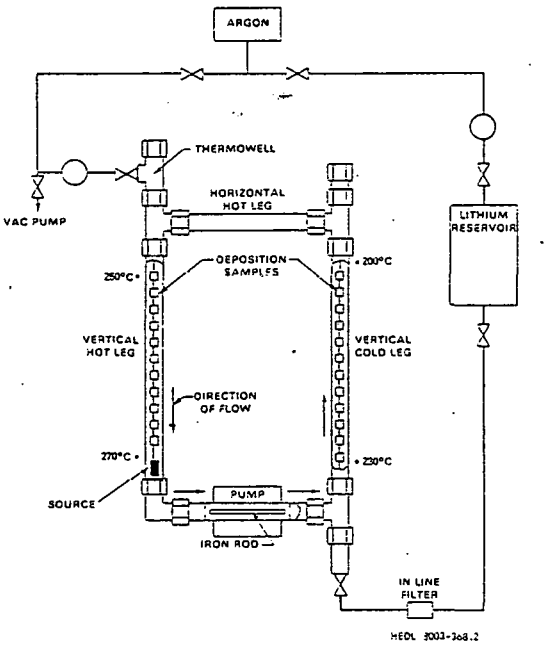

FIGURE 1. Flow Diagram of Small Test Loop

Each loop was evacuated and flushed with argon several times before lithium was introduced into the system from the reservoir. The lithium was filtered through a 60-μm filter during transfer. Nitrogen analyses of the transfer line showed only 50-ppm nitrogen. The empty loop was heated above 230°C and filled by evacuating the loop and applying 1.4×10^{5} Pa (5 psig) argon pressure to the lithium reservoir. The flow of lithium into the loop was monitored by changes in temperature and pressure. The loop was determined to be full when isothermal flow became evident after the pump was turned on. The hot leg test section increased from 250°C to 270°C along the heated, insulated section; the cold leg dropped from

230°C to 200°C along the cooling fins and exposed pipe areas. The pump was operated at very low power to maintain the desired temperature gradient. The tests were run for 950 and 1360 hours, respectively.

Lithium was removed from the loop at the end of the test duration by draining through a filter into an evacuated cylindrical tank. Typical analysis of the lithium after the test showed 170-ppm N.

To protect the loop atmosphere, the deposition samples were removed while argon was flowing through the loop. All the recovered deposition specimens were washed in de-ionized water to remove residual lithium, then dried in vacuo before weighing and counting for gamma activity. A visual inspection of the coupons was made. After exposure to lithium, all of the iron-based alloys were shiny, but the yttrium and beryllium specimens were dull and discolored. As previously noted, the nickel was severely corroded; hence, its use was discontinued.

The drained loops were analyzed for ^{7}Be every 50 mm along the tubing using a 37-mm collimated Ge/Li detector installed flush with the loop piping. The output signal from the detector was transmitted to a computer-based multi-channel analyzer. A Ge/Li detector was required since ^{65}Zn was also present on the loop walls. Zinc-65, with a gamma energy similar to ^{7}Be, had been formed during irradiation of the Cu backing plate when the ^{7}Be was made. Careful analysis was therefore necessary to determine the true ^{7}Be deposition pattern. A different backing plate is being used for future ^{7}Se preparation.

Hot Flush Procedure

After the loop was counted, it was preheated to 425°C and refilled with another volume of clean lithium from the SS reservoir. Lithium was circulated at 425°C and then drained at the same temperature. The first test took place for 6 hours and the second test was shortened to 100 minutes at 425°C before draining. Both the loop and the drained lithium were then recounted for ^{7}Be gamma activity as before.

The Beryllium-7 Experimental Lithium Loop (7BELL), illustrated in Figure 2, is constructed of 19-mm O.D. x 1.2-mm wall, type 304 stainless steel tubing. The loop, excluding the cold trap, contains approximately 4 liters of circulating lithium. A 60-Hz AC conduction electromagnetic pump circulates lithium at rates up to 0.13 l/sec and at flow velocities of 1.5 m/sec in the test section. The 7BELL operated without the cold trap on line for 1275 hours at

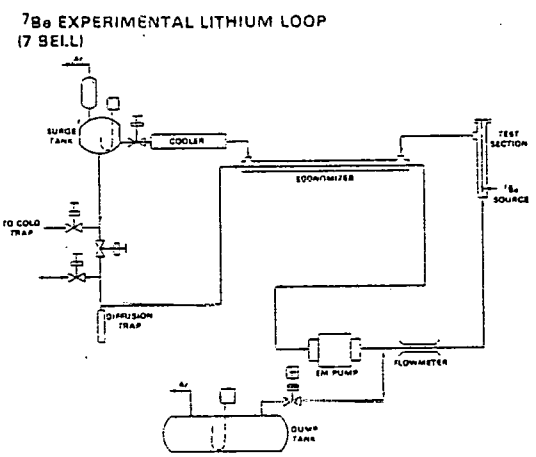

FIGURE 2. Flow Diagram of ^{7}Be Experimental Lithium Loop.

270°C hot leg and 200°C cold leg. A "diffusion cold trap," as shown in Figure 2, was part of the 7BELL circuit during the test.

RESULTS AND DISCUSSION

^{7}Be Deposition in Small Loops

A major objective of these tests was to monitor ^{7}Be deposition as a function of FMIT operating temperatures between 270°C and 200°C. Since the bottom of the vertical hot leg was held at 270°C, the ^{7}Be source was in the hottest part of the system (see Figure 1). Calculations indicate that least 90% of the ^{7}Be source was released during the test period. Before the hot flush removal procedure in both loops, much more gamma activity was detected in the 250°-270°C region than in the 230°-200°C region. (TABLE I.) This result suggests that Be does not migrate to cold areas as previously reported[3]. However, the ^{7}Be activity was not distributed uniformly throughout the loop along the horizontal and vertical sections. The activity in the horizontal hot leg was much larger than the corresponding area in the cold leg vertical section of approximately the same temperature (230°-200°C).

As shown, the lithium circulated in a counterclockwise direction. Although the ^{7}Be source was located at the bottom of the hot leg, large amounts of activity were detected in both the horizontal section and the vertical 270°-250°C test section. This result indicates that part of the material went through the cold leg and re-deposited in the horizontal and vertical hot leg regions; it did not merely deposit on the first available surface.

Hot Flush Results

After 425°C lithium was circulated in the small loops and drained, the changes in gamma activity were measured. (Table I) The cold leg area, which had very little activity after the initial test period, showed an increase in ^{7}Be since part of the ^{7}Be was redistributed by the hot flush.

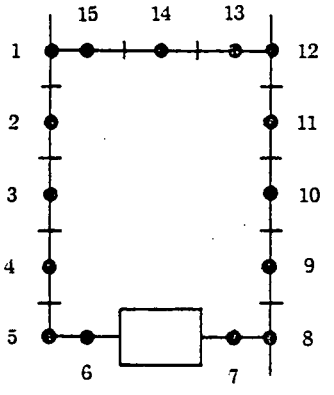

TABLE I. ^{7}Be ACTIVITY ON LOOP PIPING

Detector Location	μCi ^{7}Be Before Hot Flush	μCi ^{7}Be After Hot Flush
1	2.9	3.3
1.5	33.4	4.0
2	21.3	4.3
2.5	6.5	3.3
3	3.2	3.8
3.5	71.3	12.7
4	186.6	20.2
4.5	21.5	17.0
5	147.8	5.9
6	113.0	3.3
7	16.0	4.2
8	9.2	3.2
8.5	.9	2.7
9	2.5	3.8
9.5	.4	5.5
10	.7	7.1
10.5	2.1	8.6
11	1.1	6.6
11.5	.9	5.9
12	8.2	6.4
13	48.0	19.7
13.5	52.5	17.6
14	34.7	11.9
14.5	45.9	13.7
15	16.7	11.0

The total reduction in ^{7}Be by this procedure was calculated by the net change in activity over all measured areas. In the first test, the net reduction was 53%. In the second test, the net reduction was 76%. It should be pointed out that this reduction in gamma activity from the hot flush represents its effectiveness in removing only activity which accumulated during the actual test periods of 950 and 1360 hours of operation. On the basis of previous studies,[3] ^{7}Be penetrates into 304 SS at depths ranging from 0.54-5.33 μm after 4000 hours. It is not likely that treatment at 425°C will remove Be that had penetrated to these depths.

Analyses were made of the ^{7}Be activity remaining in the lithium drained from the loop both after 1360 hours and after the hot flush. After 1360 hours, the lithium contained 3.8×10^{-3} μCi of Be per gram of lithium. When clean lithium was circulated at 425°C and drained after 100 minutes, the concentration equalled 5.9×10^{-2} μCi of Be per gram of lithium, indicating the effectiveness of the hot flush.

^{7}Be Deposition on Test Coupons

Deposition coupons tested in the small loops included the following: Armco iron, Fe-2-1/4 Cr-1 Mo steel, cobalt, nickel, 304 SS, inert beryllium and yttrium. Yttrium specimens retained more ^{7}Be than did the other test specimens. In one test, it collected 10 to 20 times more ^{7}Be than did the 304 SS specimens at FMIT temperatures. As seen in Figure 4, there appears to be very little difference in any of the other materials tested in this temperature range.

Iron is the only material which was tested in both small loops. In the first test, preferential deposition of ^{7}Be on iron was noted. In the second small test loop, however, this result was not seen.

7BELL results

The main purpose of this test was to compare iron with stainless steel as a ^{7}Be deposition substrate at FMIT flow rates and temperatures. A 1000-hour test in 7BELL was completed with the hot leg at 270°C, and

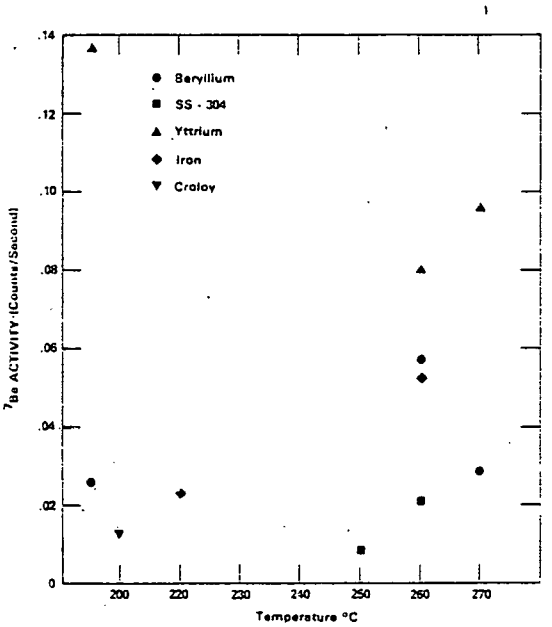

FIGURE 4. ^{7}Be Activity on Test Specimens.

the cold leg at 200°C. A flow rate of 0.10 liter/s produced a fluid boundary layer thickness similar to that in the FMIT piping. With a view towards the development of an in-line trap for ^{7}Be, preferential deposition on any material other than 304 SS was sought. However, no preferential ^{7}Be deposition on the iron was found in this case.

A diffusion cold trap was also evaluated on 78ELL as an on-line device for removing ^{7}Be. The diffusion cold trap consisted of a stagnant sump extending from the main piping system into which impurities would precipitate from the reduced temperature of the liquid. Using hydraulic conditions comparable to those anticipated in the FMIT, adequate mixing of the lithium from the main piping into the "cold finger" took place. Figure 5 shows the increased activity observed along the depth of the trap. A sharp decline in activity was noted below 150 mm, which is approximately the location

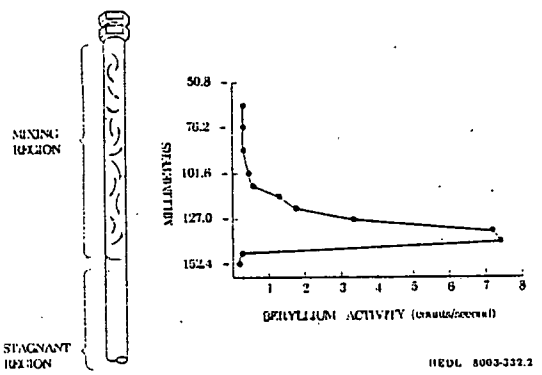

FIGURE 5. ^{7}Be Activity in the Diffusion Cold Trap.

of the solid-liquid interface measured during loop operation. The buildup of activity near this interface suggests that the diffusion cold trap shows promise as a ^{7}Be removal device.

The hot flush experiment was also performed in 78ELL. The loop was filled with lithium at 316°C and then immediately drained. Compared to subsequent procedures, the largest reduction in ^{7}Be activity was noted after this operation. This indicates that the ^{7}Be removed is primarily from the lithium film adhering to the walls, and is not from ^{7}Be diffused into the steel. Repeating the hot flush at 316°C and circulating the lithium for one hour before draining showed no additional release of ^{7}Be from the walls, even when the lithium was circulated at the maximum flow rate. The entire hot flush procedure was repeated with the lithium temperature at 425°C. Subsequently raising the temperature to 425°C released additional activity from the walls into the lithium and redistributed ^{7}Be along the piping. This result was in agreement with the expected desegregation of impurities from surfaces at higher temperatures.

CONCLUSIONS

The tests showed that part of the ^{7}Be activity in lithium is mobile and that its deposition can be controlled. Some preferential deposition on certain materials, particularly yttrium, was observed. The hot flushes were successful at removing ^{7}Be segregated to the solid-liquid interface, although they may be less successful at removing deposited and diffused activity. Continuing work will concentrate on removal of ^{7}Be in conventional hot traps at 425-450°C using yttrium and zirconium as deposition materials, and in conventional forced-flow cold traps.

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