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RECENT RESULTS ON THE GETTERING OF TRITIUM FROM MOLTEN LITHIUM*

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RECENT RESULTS ON THE GETTERING OF TRITIUM FROM MOLTEN LITHIUM

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Recent sorption experiments confirm that the mass transfer resistance in the yttrium solid phase controls the rate of tritium uptake from liquid lithium. Physical and chemical characteristics of different yttrium sorbers were measured to identify those properties that have the greatest effect on the tritium recovery rate. The BET surface areas of the five samples tested ranged from 0.005 to 0.4 m²/g; concentrations of carbon, hydrogen, nitrogen, and oxygen impurities in the sample lots increased with increasing surface area. The sample showing the highest effective sorption rate had the largest surface area but also contained the highest concentration of impurities.

1. INTRODUCTION

Tritium sorption from molten lithium using yttrium metal has been studied with a range of conditions and several different samples of yttrium [1-4]. This method has been shown to be a promising approach for the recovery of tritium from fusion reactor blankets that utilize liquid lithium as a breeding material. This approach provides an acceptable tritium equilibrium concentration and recovery rate and offers minimal introduction of impurities into the lithium. Previous papers [1,2] presented results for a single sample of yttrium which produced high sorption rates. From those data it was concluded that the principal resistance to mass transfer was in the liquid lithium and not in the yttrium sorber. Several of the factors that led to this conclusion are: 1) the magnitude of the mass transfer coefficients was in the range expected for liquid-phase resistance control, 2) consecutive sorption runs with very short separation times gave identical results indicating that the tritium concentration gradient in the liquid phase was removed with the introduction of the yttrium sample into fresh lithium, and 3) diffusivity data for hydrogen in lithium [5] and yttrium [6], when extrapolated to the experimental temperature, yielded a hydrogen diffusivity in solid yttrium that was higher than in liquid lithium. Subsequent experiments [3,4] using different yttrium samples did not achieve the high sorption rates observed initially and cast doubt on the conclusion that the liquid phase controls the mass transfer rate. Therefore, experiments have been redirected to determine the yttrium properties that affect tritium sorption rates. This paper presents physical

and chemical data on all previous sorbers and on several new yttrium samples which demonstrate that high mass transfer rates can be achieved with proper sorber preparation.

2. EXPERIMENTAL EQUIPMENT AND PROCEDURE

Yttrium sorbers were contacted with tritiated lithium in a batch contactor that has been described in previous papers [2,4]. The apparatus consisted of a liquid-metal reservoir containing dissolved tritium, a lithium transfer tank and sampling chamber, a vacuum source, and an argon supply. The tritiated lithium was drawn into the transfer tank and contacting chamber containing a yttrium sample. Yttrium sorbers were obtained from two sources: Research Chemicals (rolled slabs and powder samples) and the Oak Ridge National Laboratory Metals and Ceramics Division (sponge sample). Yttrium pellets were prepared by pressing powder samples into 6.4-mm-diam by 6.4-mm-long cylinders at 52 MPa (7500 psi) and sintering for ten minutes at 1100°C in vacuum. Yttrium pellet and sponge samples were contacted with static lithium at 300°C. A lithium control sample was removed at the beginning of each of the sorption series; the initial tritium concentration in the lithium feed reservoir was 1.10 ppm by weight. To terminate an experiment, the liquid lithium sample was removed from the contactor chamber, dissolved directly in water, and analyzed for tritium by liquid scintillation counting [7]. Yttrium samples were analyzed for common impurities. Surface areas were measured with the one-point BET method, using argon at a pressure of 15.2 KPa (114 torr) and a temperature of 77K.

Table 1. Summary of experimental results for the static tritiated lithium-yttrium sorption studies

Sample	Run No.	Sample geometry and dimensions (cm)	Sample weight (g)	Lithium temp. (°C)	Mass transfer coefficient ^a (cm/s)	Ref.
Lot 286		Slab 0.30 × 1.33 × 3.72	3.45	300	6×10^{-4}	1 ^b
		Slab 0.31 × 1.39 × 3.73	3.55	400	3×10^{-4}	2
Lot 464	Y7	Slab 0.15 × 1.33 × 3.85	3.32	300	7×10^{-6}	4 ^c
Lot 536	Y8	Slab 0.15 × 1.33 × 3.85	3.23	300	4×10^{-5}	4
	Y10	Slab 0.15 × 1.33 × 3.85	3.22	400	7×10^{-5}	4
Sponge		Irregular shape equivalent to a sphere of 1.2-diam	3.44	300	3×10^{-4}	
Pelletized powder		Cylinder 0.64-diam × 0.64	3.52	300	2×10^{-4}	

^a Mass transfer coefficients were calculated using the geometric surface areas of the sample.

^b Solid line in Fig. 1 and 2.

^c Dashed line in Fig. 2.

YTTRIUM
LOT YM-3-286

YTTRIUM
LOT Y-M-3-464C

Fig. 3. Photographs of two types of yttrium samples.

3: RESULTS AND DISCUSSION

Figure 1 shows the results of the batch sorption experiments for new yttrium sponge and pellet samples at 300°C, where the ratio of tritium concentration, C , and the initial tritium concentration, C_0 , is plotted as a function of the contact time. The line in Fig. 1 represents the highest sorption rate, reported by Clinton and Watson in an earlier study [1]. The new data follow the initial steep slope of the early experiments. Figure 2 shows similar results for previous experiments using the three sample lots of rolled yttrium, which gave poorer performance. Considerable variation in the sorption rates of the different samples of yttrium is apparent from Fig. 2. Table 1 sum-

marizes the experimental results for the samples used in sorption studies to date. The mass transfer coefficients were calculated as if sorption was a first-order process; this was done to allow comparison of the sorption rates, but not to suggest a mechanism or rate expression.

Since the nature of the solid phase appears to determine mass transfer rate, it has become important to characterize the yttrium sorbers. Physical and chemical analyses of the three lots consisting of rolled samples, sponge, and pellet sorber material were conducted to investigate the role of structure and purity in tritium sorption ability. Photographs of these materials are shown in Figs. 3 and 4. The original yttrium sample (lot 286) that gave the best performance was dark gray and brittle, with a swollen and laminated structure. Data of Clinton and Watson from lot 286 are shown in Figs. 1 and 2 as the solid curve. The newer rolled samples of yttrium (lots 464 and 536) were ductile, shiny, solid, and nonporous.

Fig. 1. Batch sorption results for yttrium sponge and yttrium pellet at 300°C, plotted with a previous static sorption experiment by Clinton and Watson [1].

Fig. 2. Comparison of batch sorption results for rolled yttrium samples at 300 and 400°C.

YTTRIUM SPONGE

YTTRIUM METAL POWDER

LOT Y-M-3-648C

Pressed at 7500 psi

Sintered in vacuum at 1100°C for 10 min

Fig. 4. Photographs of yttrium sponge and pelletized powder samples.

Data from these samples (Fig. 2) show sorption rates much slower than the rate for lot 286. A sample from lot 464 was used for run Y7 and from lot 536 for runs Y8 and Y10. Yttrium sponge has the appearance of a relatively porous, metallic stone, and the pelletized powder is quite porous. These samples (see Fig. 1) gave mass transfer rates comparable to lot 286.

Chemical analyses for impurities in the three lots of rolled yttrium and the sponge sample used for sorption experiments are shown in Table 2. The pellet sample was not analyzed for impurities but was fabricated from high purity powder. As can be seen, the yttrium sample lot containing the highest concentration of impurities (lot 286) gave the best mass transfer results. Because lot 286 contained

815 ppm of hydrogen, while lot 536 had only 3 ppm, it was suspected that the favorable mass transfer data might be due to a hydrogen isotope exchange rather than sorption. To test this, sample 536 was loaded with 800 ppm of hydrogen. Sorption runs on this sample, however, showed an insignificant reduction in tritium concentration in the lithium, proving this hypothesis incorrect.

Table 3 shows the surface areas and relative densities of the various yttrium samples. Lot 286 has more surface area than lot 536 by two orders of magnitude. The yttrium sample of lot 536, which has the lowest concentration of impurities and the smallest surface area, is closest to the theoretical density of yttrium (4.5 g/cm³). Micrographs of the three rolled samples of yttrium (Fig. 5) show the effect of

Table 2. Chemical analysis results for yttrium samples

Sample lot	Impurity concentration in yttrium					
	C (ppm)	H (ppm)	N (ppm)	O (%)	Ta (%)	Fe (%)
286	1727	315	1644	4.0	0.1	0.03
464	805	69	320	0.7	0.1	a
536	316	3	114	0.5	0.1	a
Sponge	b	200	180	0.45	a	a

^aNone detected.

^bNot determined.

Table 3. Physical data from analysis of yttrium samples

Sample	Surface Area (m ² /g)	<u>Density of Sorber</u> <u>Density of Yttrium</u>
Lot 536	0.0051	0.982
Lot 464	0.034	0.968
Sponge	0.055	0.867
Pelletized powder	0.090	0.505
Lot 286	0.368	0.503

YTTRIUM METAL

LOT 464

LOT 536

LOT 286

Fig. 5. Micrographs of sample lots of rolled yttrium.

oxygen impurity on the production of surface area. Oxygen appears as isolated dots in lot 536; the larger amount of oxygen in lot 464 appears as striations, and the oxygen level in lot 286 is so high that sheets formed by rolling delaminated, producing a relatively high surface area.

The highest sorption rates have occurred with yttrium samples having both high surface areas and impurity concentrations. It is suspected that surface area is the more important parameter, but further experiments will be necessary to confirm this. Bulk yttrium sample analyses gave impurity levels, but the degree that the impurities are concentrated on the sample surface will have to be determined if impurity levels are found to be important to tritium sorption. However, these limited data have shown that high impurity concentrations do not necessarily inhibit tritium sorption rates and may even increase them.

4. CONCLUSIONS

Gettering of tritium by solid yttrium appears to be a feasible method for tritium recovery from liquid lithium. Recent sorption experiments with yttrium sponge and pellet samples confirm that the solid phase controls tritium mass transfer rates. Hence, the physical and chemical properties of yttrium samples strongly affect their sorption rates for tritium from molten lithium, with high surface areas and porosities needed to achieve acceptable rates. Since samples with high surface areas also had high impurity levels, the effects were not separable. Impurities have not been found to have an adverse effect. A more complete understanding is needed to evaluate the practicality of this processing technique. Preparations are being made to use sorbers with higher surface areas and investigations continue concerning the effects of impurities, sorber surface area, and porosity on the rate of tritium sorption.

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