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Summary Report for Zinc 65 Contamination Control

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Summary

Radioactive zinc, ^{65}Zn, was detected after extraction of 215 TPBARs in from TVA reactor fuel cycle 6. A team consisting of Tritium Engineering, Tritium Operations, Tritium Radiation Control, and Savannah River National Laboratory personnel evaluated the risk and response and developed short, medium and long term goals for contamination control. One of the goals was incorporated into site Performance Based Incentive CO 3.4, to optimize the filter geometry and operating conditions for the Tritium Extraction Facility. This goal included a scoping study to determine if the contamination could be contained within the high radiation environment of the furnace module as well. In order to optimize the filters studies were conducted to independently evaluate the effect of pore size on pumping efficiency and zinc trapping efficiency (1). A study was also conducted to evaluate the effect of temperature on the trapping efficiency and adhesion (2). In addition, the potential for chemically trapping zinc in the lithium trap was evaluated using a thermodynamic study (3) followed by preliminary experimental testing (4).

Based on the work that was completed it is determined that a 20μm filter heated to between 120 and 200°C will act as an effective physical trap for zinc vapors. It may be possible to chemically react zinc with copper or cobalt to form zinc intermetallic compounds or alloys but additional work under more prototypic conditions are required.

Background

Radioactive zinc 65 (^{65}Zn) has been extracted from irradiated TPBARs at levels that produce a signature greater than background (5). Review of the potential sources of ^{65}Zn from the TPBAR sub-elements indicates that it is an activation product of natural zinc and that it is present as a low level contaminant in several of the TPBAR materials (6). The results from the initial post-mortem examination of the piping removed from the TEF, and a literature review of vapor phase deposition indicated that certain conditions may be beneficial to the growth of the zinc whiskers. Consequently 20 μm filters heated to 200°C were installed in key areas of the TEF process lines (7). The subsequent post-mortem examination of the filters (8), especially gamma scans, indicated that ^{65}Zn had been trapped within these locations, although the deposit morphology was difficult to characterize due to the presence of native Zn on the filters (9). These results lead TEF to sponsor this effort to optimize filter capturing and evaluate the possibility of capturing zinc within the lithium trap.

An experimental program was initiated to develop and validate conditions that will effectively trap Zn vapor released during extraction in the TEF. The work was divided into three experimental tasks and a literature/thermodynamics study. The first experimental task was to determine the effectiveness of various pore sizes of filter elements (1) which revealed that 20 μm

filters would be effective to trap the zinc vapor. The second task was to determine the effect of filter temperature on zinc vapor deposition (2) which resulted in a recommendation that the filters be heated to between 120 and 200°C. The thermodynamics study (3) indicated that copper and cobalt may be effective at trapping zinc in a chemical form while avoiding forming hydrides. If it is possible to capture the contaminants chemically in the lithium trap, then the contamination would remain in the high radiation area, which would reduce potential dose to the worker. The final experimental task was to determine whether the zinc vapors could be chemically bound at conditions typical of the lithium trap (4). The overall approach for capturing the zinc vapors is described in Ref. 9. This report summarizes the results of the experimental testing.

Results

Figure 1 shows the effect of inserting filters with different pore sizes in the pumping train. It is apparent that filters with pore diameters of 5 to 20 μm have little to no effect on the pumping speed and smaller pores adversely affect pumping times.

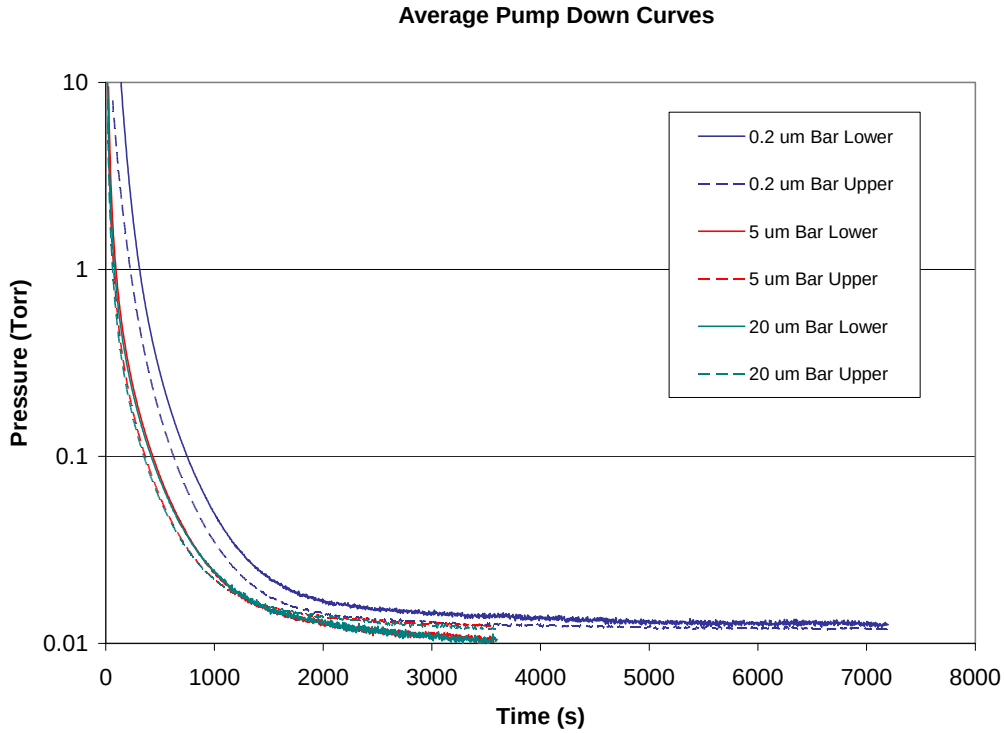

Figure 1. Effect of pore size on pumping times.

The effect of pore size on zinc trapping efficiency was determined by using thermal evaporation of zinc in a glass and metal system. The system was pumped with a scroll pump that would pull the chamber to pressures of 50 to 90 millitorr prior to heating either the zinc containment vessel or the filter holder. The gravimetric data from these experiments are listed in Table 1. There is no apparent difference in the efficiency of zinc vapor trapping due to the amount of scatter in the data.

Table 1. Test Condition and results

Filter	Vessel Temp (°C)	Time T>300°C (s)	Δ mass (mg)	Comments
0.2 µm – 1	333	3965	0.40	Old Config.
20 µm – 4	340	3954	1.31	Old Config.
0.2 µm – 2	330	7492	2.89	New Config
1 µm – 1	325	7613	1.73	New Config used Zn
5 µm – 1	320	6613	2.57	New Config used Zn
10 µm – 1	324	7689	1.49	New Config used Zn
20 µm – 6	325	7546	1.82	New Config used Zn
20 µm – 7	350	8169	2.14	New Config New Zn

With the 20 µm pore size selected, an optimized temperature was sought. The temperature ranged from 60 to 200°C. The low temperature was selected based on the current heat trace temperature used for the piping while the upper temperature was based on the temperature limit of the glovebox. Several criteria were used to select the optimized temperature condition. The temperature was to provide the most efficient temperature for zinc capture and also for zinc retention. The typical deposit on the 20µm filters held at 60, 120, and 200°C is shown in Figure 2. The deposit is more prevalent on the surface of the filter tested at 60°C, but this deposit was the easiest to remove based on the simple adhesion test that used.

(a) **(b)** **(c)**

Figure 2. Appearance of zinc deposit on 20 µm filters at (a) 60, (b) 120 and (c) 200°C.

The deposition weights and adhesion characteristics are listed in Table 2. These results suggest that 60°C is more efficient at trapping the zinc. This result is valid, except that the zinc is poorly adherent to the surface with approximately twice as much material being removed from the 60°C sample compared to the 120 and 200°C. The concern with poorly adherent material is that contamination could spread to other parts of the glovebox during maintenance and other activities.

Table 2. Test Condition and results

Test ID	Date	Vessel T (°C)	Filter A (°C)	Filter B (°C)	Δ mass (mg)	Δ loss ad. (mg)
60-1	1-26	340	55	52	3.8	0.8
60-3	2-10	380	57	50	2.1	0.5
60-4	3-3	384	56	50	3.7	0.5
120-2	2-28	340	116	96	1.6	0.1
120-4	3-1	384	116	97	2.9	0.2
200-1	1-26	340	196	160	2.6	0.3
200-2	2-8	340	200	165	5.5*	0.3
200-4	2-28	384	199	149	2.4	0.2

Notes: "A" refers to thermocouple mounted Above the filter and "B" to the one below. "ad" refers to mass loss during tape adhesion test. *sample was subjected to a double exposure at 400°C. Missing Test ID are due to poorly executed experiments. The thermocouples were reset on Feb 10, 2011 which resulted in changes in measured vessel temperatures.

Thermodynamic calculations and experiments were conducted that showed that copper and cobalt would capture zinc as compounds without also acting as hydrogen getters. Most materials that had favorable energy of formation for zinc also had a propensity to act as hydrogen getters as well. Figure 3 shows the results of a thermodynamic calculation for zinc and hydrogen in contact with yttrium. This calculation indicates that both zinc and hydrogen would be captured on the yttrium. A further examination of the Y-H diagram indicates that the temperature that hydrogen would be released is over 850°C which is well over the conditions in the lithium trap.

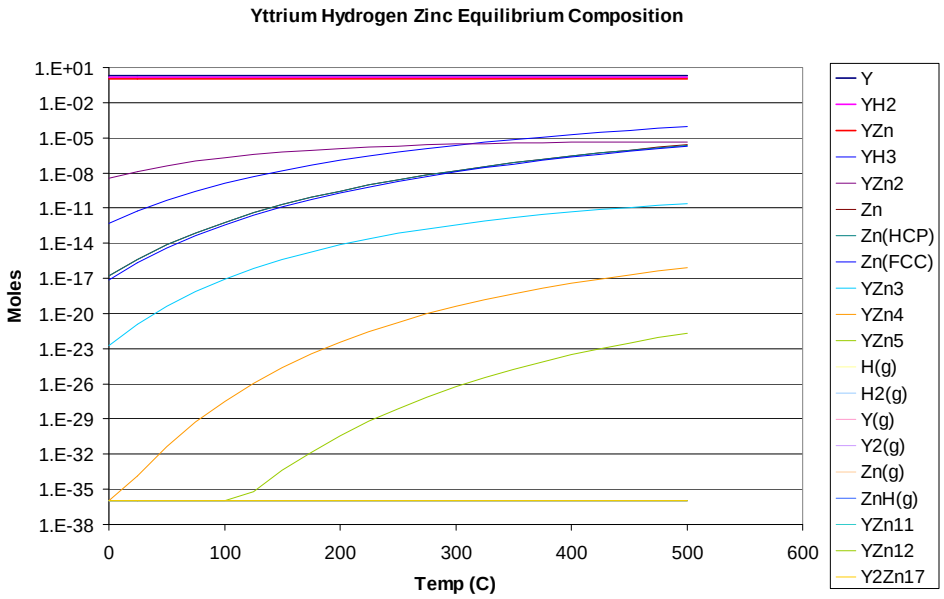

Figure 3. Thermodynamic calculation showing the resultant composition for the Yttrium - Hydrogen - Zinc system.

A scoping study was conducted to determine if the calculations and literature review could be proven experimentally. Both copper and cobalt components were tested. Zinc was deposited on the copper components but the zinc did not react with the copper, as shown in Figure 4.

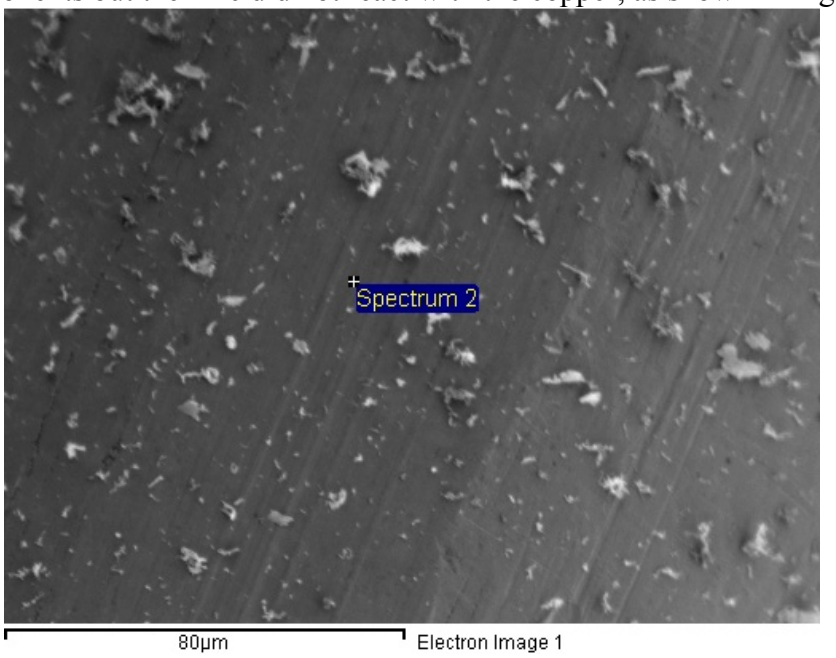

Figure 4. Copper substrate after 450°C exposure in zinc vapor.

As can be seen the zinc is present as discrete deposits. Analytical techniques, scanning electron microscopy and x-ray energy dispersion spectroscopy demonstrated that the zinc and copper did not alloy. However, this sample was exposed to a higher shell temperature than what was used for vaporization, yet had a zinc deposit. This result suggests that trapping in the lithium basket may be possible even without chemical binding. Additional work using prototypic conditions, i.e., temperature and pressure of the lithium basket, is needed to validate this possibility.

Conclusions

Using a filter with a pore diameter of 20 µm and holding it at temperature between 120 and 200°C will provide a zinc deposition surface that is efficient for trapping in addition to adhesion.

Additional work is needed to develop a suitable substrate for zinc deposition in the lithium trap. Capture of zinc in the lithium trap would be beneficial so that the gamma producing zinc remains in the radiation area rather than being deposited in a glovebox that is designed for beta radiation. In addition, the trap can be disposed of with the TPBARs rather than creating a new waste stream.

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