

Testing of New Antimony Selective Media SbTreat at Loviisa NPP, FINLAND – 14241

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ABSTRACT

SbTreat is a new inorganic antimony selective media. It was tested for the removal of antimony, both non-radioactive and radioactive (^{124}Sb), from the outgoing sealing water of the Primary Main Coolant Pumps (PCP) at Loviisa NPP (VVER-440, Finland). The media was utilized in powdered form in a special 10-inch cartridge. PCP output water contained ^{124}Sb and stable antimony in the level of 10 kBq/L and 2 ug/L, respectively. The feed (rate 5-10 gph) to the cartridge was initiated during the start-up operations of the Lo1 Reactor in September 2012. The first SbTreat cartridge (SbTreat dose 300 g) removed ^{124}Sb with a DF of 100 initially. The maximum DF for non-radioactive antimony was about 40. The first cartridge was exhausted after 6030 L of PCP water had been treated. The second SbTreat cartridge performed clearly better than the first one, maintaining DF 100-10 for ^{124}Sb until 9000 L of PCP water had been treated.

INTRODUCTION

Radioactive antimony ($^{122, 124, 125}\text{Sb}$) may exist in the PWR and BWR water circuits and waste streams in activity concentrations comparable to that of radiocobalt ($^{57, 58, 60}\text{Co}$). Radioactive antimony is a neutron activation product of stable antimony and may give a major radiation dose to the maintenance personnel during the NPP service shutdowns. Standard water treatment systems (demineralizers, filters, etc.) are generally inefficient for the removal of radioactive antimony [1, 2]. However, several new ion selective media have been developed recently, capable of extremely high removal of $^{124, 125}\text{Sb}$ from NPP primary circuit and floor drain waters [2-5].

SbTreat is a new zirconium oxide based material under development by the University of Helsinki and Fortum that has been tested for the removal of ^{124}Sb from the Primary Coolant Water (PCW) from Loviisa NPP (VVER-440, Finland) [5]. Laboratory-scale column experiments conducted using granular (grain size 0.30-0.15 mm) SbTreat and PCW showed very high decontamination factors (DF) up to 30,000 for ^{124}Sb . An improved product format of SbTreat, a finely divided powder allowing high flow rates has also been developed and tested recently [6].

EXPERIMENTAL

A pilot scale test has been conducted to remove stable and radioactive antimony from the outgoing sealing water of the Primary Main Coolant Pumps (PCP) during the Loviisa 1 (VVER-440, Finland) reactor restart in September 2012. For the experiments, SbTreat powder (about 0.3 kg) was formulated and packed in 10" FlipFilter cartridges by Graver Technologies Inc. (Glasgow, DE, USA). The SbTreat cartridge (Fig.1) was fed by a sampling line from the PCP output water at a flow rate of 20-40 L/h. Initially the PCP output water contained ^{124}Sb and stable antimony in the level of 10000 Bq/L and 2 ug/L, respectively. During the startup procedures the major chemical constituents were boric acid (14 g/L) and ammonia (range 20-25 mg/L).



Fig. 1. SbTreat Pilot Test Facility. SbTreat cartridge housing (lower right) was fitted with radiation shielding.

RESULTS

The feed of PCP water in the first SbTreat cartridge was initiated on September 18, 2012. The cartridge removed ^{124}Sb with a decontamination factor (DF) of 100-10 until about 2500 L of water had been treated (total ^{124}Sb removal at this point 93 %) (Fig. 2). The DF's for stable Sb were somewhat lower (DF = 40-4). After this the DF for ^{124}Sb fell below 10 and the cartridge was exhausted for ^{124}Sb (DF < 1) when 6030 L of water had been treated. For the non-radioactive antimony, the exhaustion took place somewhat earlier at 5500 L.

After the exhaustion of the first cartridge, the second SbTreat cartridge was installed online. At this time, the feed activity of ^{124}Sb was still at the level of 10000 Bq/L. The feed concentration of the non-radioactive antimony had fallen close to, or below the detection limit (0.1 µg/L). Thus the DF's measured contained a large error. The second SbTreat cartridge performed clearly better than the first one, the DF 100-10 for ^{124}Sb could be maintained until about 9000 L of water had been treated (Fig. 3). Laboratory tests show that an increasing boric acid concentration decreases the antimony uptake in SbTreat. During the use of the second cartridge, the concentration of boric acid in the feed was lower (5-6 g/L) than during the operation of the first cartridge. Thus the most likely cause for the better performance of the second cartridge is the

lower boric acid concentration in the feed.

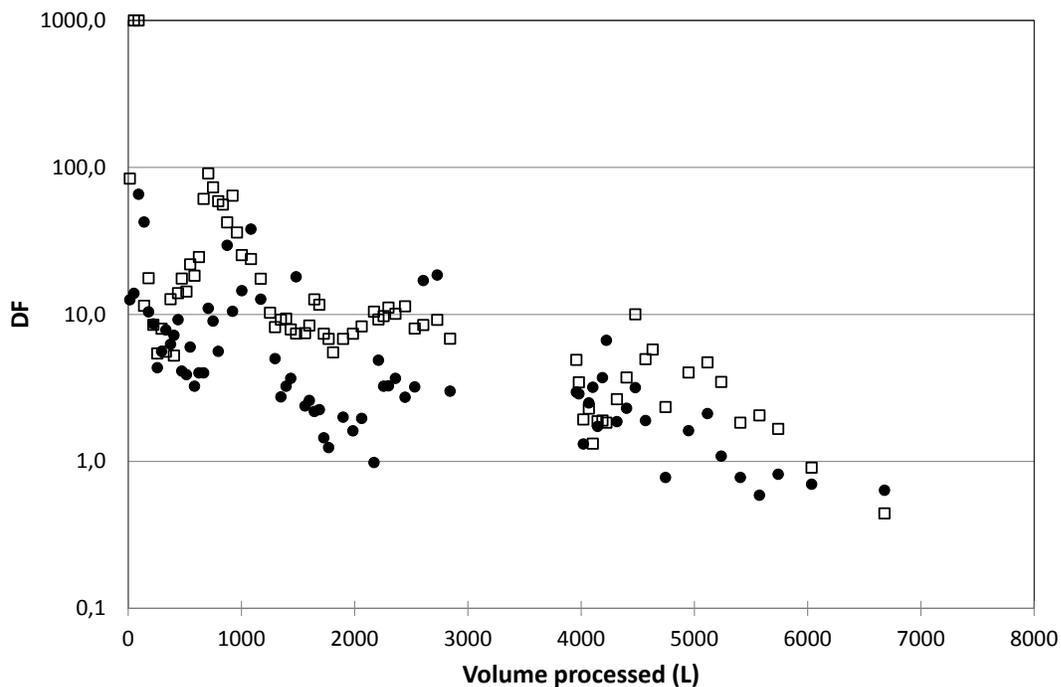


Fig. 2. Decontamination factors (DF) for ^{124}Sb (\square) and for non-radioactive antimony (\bullet) for the first SbTreat cartridge.

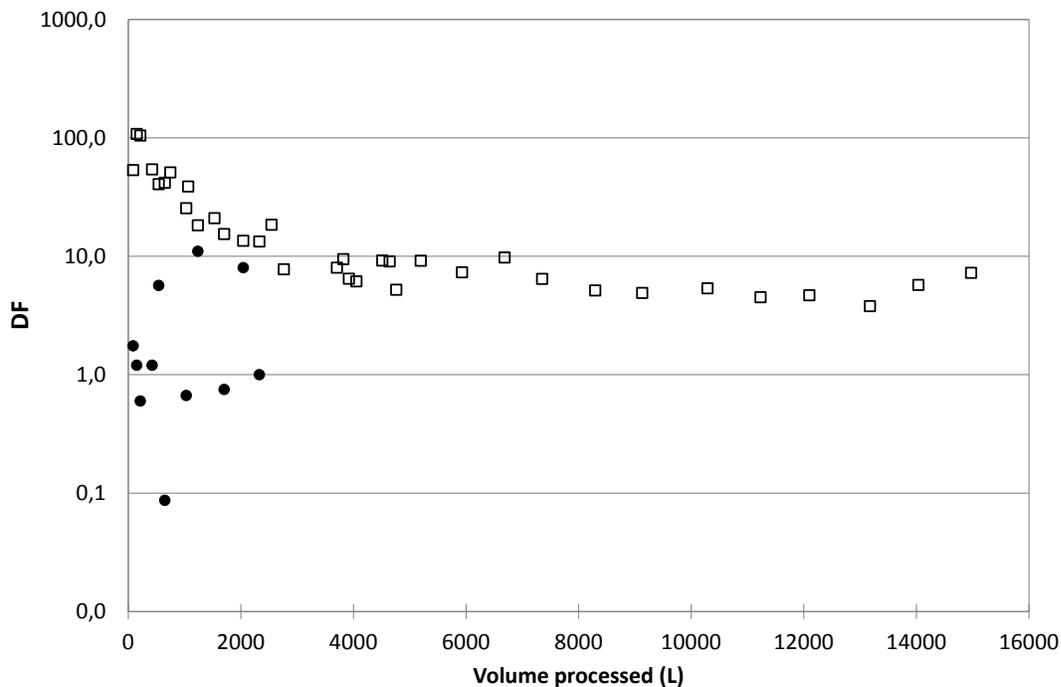


Fig. 3. Decontamination factors (DF) for ^{124}Sb (\square) and for non-radioactive antimony (\bullet) for the second SbTreat cartridge.

The decontamination factors for ^{124}Sb were clearly higher than for non-radioactive antimony. Antimony can exist in solution in two oxidation states (+3,+5) and in several hydroxyl species (e.g. $\text{Sb}(\text{OH})_6^-$, $\text{Sb}(\text{OH})_3$ (aq), $\text{Sb}(\text{OH})_4^+$), depending on the pH and redox conditions. The difference in DF*s is most likely explained by the different chemical speciation of radioactive and non-radioactive antimony.

The DF's were however clearly lower than in earlier small-scale tests where powdered SbTreat was utilised in small filtration capsules [6]. This may be a scale-up effect or may be due to the higher relative flow rates that were used in these latest tests.

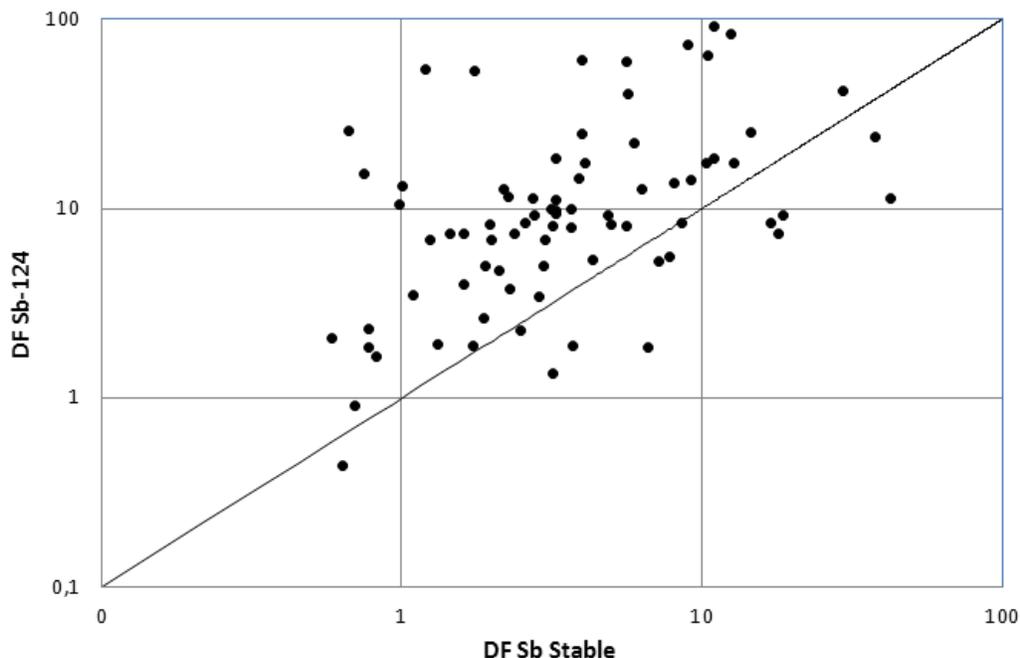


Fig.4. Decontamination factors (DF) of stable antimony vs. DF of ^{124}Sb

CONCLUSIONS

In terms of water volume processed per unit mass, the SbTreat cartridges (0.3 kg SbTreat) had good processing capacities for the removal of ^{124}Sb , 8300 L/kg and 30000 L/kg. However, the decontamination factors were only modest and much lower than what had been observed in earlier tests. Further work is needed to optimise the operating conditions to improve the decontamination factor.

REFERENCES

1. Electric Power Research Institute, Analysis of Advanced Liquid Waste Minimization Techniques at a PWR, TR-109444, 1998.
2. R. Harjula, A. Paajanen, R. Koivula, E. Tusa and R. Kvarnström, Removal of Antimony-124 from PWR Coolant Water, Proceedings of Waste Management 2009 Conference, March 1 – 5, 2009, Phoenix, AZ.
3. P.Y. Yarnell and A. Tavares, Ion Exchange media for the Reduction of Liquid Radwaste at Commercial Power Plants, Proceedings of Waste Management 2008 Conference, February 24-28, 2008, Phoenix, AZ.
4. P.Y. Yarnell, Testing of Antimony Selective Media for Treatment of Liquid Radwaste, Proceedings of Waste Management 2007 Conference, February 25 -March 1, 2007, Tucson, AZ,
5. R. Harjula, A. Paajanen and R. Koivula, Removal Of Sb-125 And Tc-99 From Liquid Radwaste By Novel Adsorbents, Proceedings of Waste Management 2006 Conference, February 26 – March 2, 2006, Tucson, AZ.
6. R. Harjula, A. Paajanen, R. Koivula, I. Välimaa, E. Tusa, M. Pehkonen and R. Kvarnström, Development and Testing of New Antimony Selective Media, Proceedings of Waste Management 2011 Conference, February 27 - March 3, 2011, Phoenix, AZ.