



Contribution ID: 81

Type: Verbal

Removal of antimony-124 from PWR coolant water

Selective ion media, e.g. inorganic adsorbents and ion exchangers, are increasingly used for the removal of key radionuclides such as ^{60}Co , ^{90}Sr and ^{137}Cs from nuclear waste effluents due to their radiation stability, high processing capacity and high decontamination efficiency [1,2]. The materials used that are commercially available (e.g. zeolites, titanates, silicotitanates, hexacyanoferrates) are cation exchangers or adsorbents, which can remove efficiently cationic and neutral radioactive species. Inorganic anion exchange materials are quite rare and do not possess high selectivity.

Considering radiation doses to personnel and environment, $^{58,60}\text{Co}$ and ^{137}Cs are the most critical radionuclides in nuclear power plant (NPP) waste liquids and water streams. Improved processing systems have been able to reduce markedly the discharges of these radionuclides at many utility sites and further efforts have been directed to remove other radionuclides such as ^{51}Cr , ^{110m}Ag and ^{125}Sb that dominate in solution after cesium and cobalt elimination. Much attention has been paid recently to ^{125}Sb . It may exist completely in soluble form in the Floor Drain Waters [3]. In solution, antimony can exist 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. These chemical features indicate that it is difficult to remove antimony from solution.

Recent tests showed that standard demineralizer resins and ion selective media are ineffective for the removal of Sb from liquid radwaste [4]. However, some commercially available inorganic cation exchangers, such as CoTreat, can remove ^{125}Sb from NPP Floor Drain water with good efficiency [3] in some cases, but their utilization is obviously restricted to cationic antimony species. Regarding other methods, chemical additives coupled with ultra filtration has been shown to be effective method for ^{125}Sb removal in a test program conducted at Duke Power Company's Oconee plant [5]. Study of other methods such as electro-deionization and hollow-fibre filtration is underway e.g. in the EPRI Low-Level Waste program [6].

In Loviisa NPP (PWR, Finland) about 50 % of the radiation dose received by personnel during the service shut-down period is caused by ^{124}Sb . Immediately after shut-down, ^{124}Sb is released from fuel into the primary coolant water at levels greater than 100,000 Bq/L ($> 3 \times 10^{-3}$ Ci/mL).

The mixed-bed organic resin demineralizer system used for activity removal under routine reactor operation is inefficient in removing the released ^{125}Sb . Loviisa NPP has thus established a project to study and assess various ^{124}Sb abatement technologies.

As a part of this project, Laboratory of Radiochemistry has just initiated a laboratory study to remove ^{124}Sb from the primary coolant water of Loviisa NPP using novel zirconium oxide (ZrO_2) sorbents [7]. Samples for the tests were obtained from service shut-down of Loviisa Unit 1 that started August 8, 2008. Analysis of water samples showed that they contained soluble ^{124}Sb (after 0.45 μm filtration) at level of 600,000 Bq/L. Static uptake test showed a high uptake of ^{124}Sb b

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Track Classification: Chemistry of Nuclear Fuel Cycle, Radiochemical Problems in Nuclear Waste

