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Conversion of I₂ in ionic forms on the composite materials "Fizkhimin"™ in water coolant at nuclear power plants

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It is known that the activity of radioactive iodine in the water coolant increases dramatically during the shutdown of reactors on the Russian working WWER- or RBMK-type NPPs. For example, the radioactivity of ¹³¹I in the water coolant increased 30 to 200 times during the shutdown of the reactor on the RBMK-type NPP such as the Leningrad and Smolensk NPPs. One of the reasons of this activity rise is that during the shutdown of a reactor the temperature of the fuel inside the fuel elements drops, which results in the reduction of the fuel volume and the appearance of free space inside the fuel elements. Simultaneously, water from the coolant penetrates into the fuel elements through defects in the fuel cladding and dissolves radioactive iodine and caesium accumulated in the gap between the fuel and cladding during normal operation of NPP. The dissolved radioactive iodine and caesium go as ions to the water coolant. Due to different redox reactions in a water coolant radioactive iodine can be present as ionic (I⁻, IO₃⁻) and molecular (I₂) species. Furthermore, the radiation-induced destruction of the ion-exchange resins can lead to the formation of organic species (such as CH₃I) in a water coolant. At the same time the content of radioactive iodine in the gas phase increases by more than an order of magnitude, because molecular and organic forms of radioactive iodine make a major contribution to the pollution of the gas phase since their factors of distribution between the water and gas phases are several orders of magnitude higher than those for ionic forms. We suggest that an analogous increase in the radioactive iodine content in the water coolant and gas phase during the shutdown can occur also in the currently working and new developed NPPs using a water coolant.

To decrease the radionuclide content in the water coolant, cation- and anion-exchange resins are usually used during a normal operation of NPPs. For example, cation-exchange resin KU-2 and anion-exchange resin AV-18 were used on the Russian NPPs for coolant decontamination. However, anion-exchange resins can absorb effectively only ionic forms of radioactive iodine and possibly I₂ as anion I₃⁻.

We studied the sorption of CH₃¹³¹I and I₂ on an AV-18 ion-exchange resin from the water coolant which takes places on the some of the Russian WWER-type NPPs. It was found that the AV-18 anion-exchange resin can absorb I₂ and CH₃¹³¹I from the solution. After 2 h of contact between liquid and solid phases, I₂ absorption was about 70%, whereas the CH₃¹³¹I absorption was only about 30.0%. Anion-exchange resin AV-18 effectively absorbs I₂ (more than 99%) from this water coolant of a WWER-type NPP only after 24 h of contact between the liquid phase and the resin. In this case the distribution factors K_d for I₂ were higher than 103 cm³/g, whereas the distribution factors K_d for CH₃¹³¹I were lower than 200 cm³/g.

Because of a low rate of sorption of I₂ and CH₃I the anion-exchanged resin AV-18 could not effectively and quickly localize these forms of radioactive iodine from the water coolant. These findings made it apparent that a new more effective material to absorb molecular and organic iodine from water solutions was required, and it was desirable to employ materials used in working NPPs to coolant decontamination.

In our experiments, we used ion-exchange resin KU-2, which we modified to obtain sorptive materials. The materials IPCE-C1 and IPCE-C2 with about 8.0 and 5.5 wt.% of Cu, and also IPCE-CS with about 10.1 wt.% of Cu and Ag were obtained and used for our bath experiments.

For these materials, we studied sorption of different forms of radioactive iodine (¹³¹I⁻, ¹³¹IO₃⁻, CH₃¹³¹I) and I₂ from water coolant which takes places on the some of the Russian WWER-type NPPs.

Preliminary experiments showed that the non-modified cation-exchange resin KU-2 has a low sorption capac-

ity for CH3131I and I2 in the studied solution: the distribution factors are not higher than 5.0 cm³/g at V/m = 100.

At the same time under similar experimental conditions the distribution factors K_d for I2 for all developed materials are higher than 103 cm³/g. The distribution factors K_d for ionic forms change from 20.0 to 150 cm³/g, whereas for the organic form, they are not more than 11.0 cm³/g.

As follows from the data on I2 sorption kinetics on our developed materials and anion-exchanged resin AV-18, during 15 min of contact between the solid and liquid phase practically full I2 absorption (more than 99.0%) takes place on the modified resin KU-2 vs. 50% on the resin AV-18. As above-mentioned, the 99% absorption of I2 on anion-exchange resin AV-18 can be achieved only after 24 h. Unfortunately, the developed materials have low efficiency for CH3131I in the studied solutions.

So, our findings allow us to conclude that using a combination of our developed materials and anion-exchanged resins such as AV-18 would allow to more effectively decontaminate the water coolant from ionic and molecular forms of radioactive iodine and partially organic form in working and new developed NPPs.

Also note that the application of the new materials will not require changes in either the designs of NPPs or the reactor's water coolant decontamination sites and will allow to bring down the load on the iodine filters during the shutdown and refueling of a reactor, as well as normal operation of the NPPs.

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