

Development of polymeric adsorbents for recovery of uranium from seawater

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Extraction of uranium from unconventional resources, where uranium is in low concentrations as in seawater, can be orders of magnitude higher in cost than extraction from conventional sources. As a part of the Fuel Cycle Technology Research and Development Program in the United States Department of Energy, Office of Nuclear Energy, Oak Ridge National Laboratory (ORNL) is developing new adsorbents with higher capacities, selectivities, and durability for the cost effective extraction of uranium from seawater, the most challenging but highest-payoff unconventional resource.

Over the last three years, the key focus of the ORNL R&D efforts has been on increasing the adsorption capacity of amidoxime-based polymeric adsorbents by the radiation-induced graft polymerization on high surface-area polyethylene fibrous trunk materials. These trunk materials have been fabricated through an "islands-in-the-sea" fiber-spinning method, which can considerably enhance the surface area of the high-density polyethylene fibers without compromising its mechanical properties. Acrylonitrile and methacrylic acid can be effectively grafted onto these high surface-area fibers followed by conversion of the nitrile groups to amidoxime groups. Marine testing of these poly(acrylamidoxime-co-methacrylic acid) adsorbents at the Pacific Northwest National Laboratory Marine Sciences Laboratory showed uranium adsorption capacities, for the extraction of uranium from seawater, that were more than three-times higher than that previously reported. We are continuing to work to increase the adsorbent capacities of the amidoxime-based adsorbents through optimization of the polymerization conditions and investigation of new grafting methods without the use of ionizing radiation such as Atom-Transfer Radical Polymerization. We have also successfully manufactured several braided fiber adsorbents that are potentially suitable for marine deployment.

In addition to these studies, we have also explored methodologies to develop novel adsorbents that contain chelating sites with higher uranium binding affinity and selectivity than that exhibited by the current amidoxime-based adsorbents. Using a combined theoretical and experimental approach, it was discovered that amidoximate anion binds uranyl ion in the $\text{O}2$ binding motif. Building on this knowledge, de novo structure-based design methods were used to design bis(amidoxime) ligands in which the ligands can engage the uranyl ion in the most stable binding motif. The best bis(amidoxime) ligands were synthesized and experimentally found to exhibit a log K for uranyl complexation that were at least 4 to 8 orders of magnitude stronger than the amidoxime ligand. Moreover, these bis(amidoxime) ligands were significantly more stable to acid stripping of the adsorbed metals...

see attachment for full abstract.

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