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RESIN-BASED PREPARATION OF HTGR FUELS: URANIUM LOADING DEVELOPMENT STUDIES*

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*Research sponsored by the Energy Research and Development Administration under contract with the Union Carbide Corporation.

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The reference fuel kernel for recycle of ^{233}U to HTGRs (High Temperature Gas-Cooled Reactors) is prepared by loading carboxylic acid cation-exchange resins with uranium and carbonizing at controlled conditions. The carbonized products must be spheres with high U contents containing only U, C, and O as major constituents. Uranium loading flowsheet conditions were investigated and demonstrated in engineering equipment.

A resin and process condition to give acceptable loaded spheres was initially developed using the hydrogen form of the cation resin and UO_3 to maintain acid-deficient uranyl nitrate.[1] The purified $^{233}UO_2(NO_3)_2$ solution from a fuel reprocessing plant contains excess HNO_3 (NO_3^-/U ratio of about 2.2). Considering the requirements for remote operation, accountability, and control of nuclear criticality, the usual processes for preparing UO_3 and the in-cell use of UO_3 did not seem acceptable. Therefore, the amine extraction process described below was developed.

The reference flowsheet for a ^{233}U recycle fuel facility at Oak Ridge uses solvent extraction of nitrate by a 0.3 M secondary amine in a hydrocarbon diluent to prepare acid-deficient uranyl nitrate (Fig. 1). This nitrate extraction, along with resin loading and amine regeneration steps, was demonstrated in 14 runs[2] using components and procedures developed as part of sol-gel studies.[3] All of the resin loading tests were completed as planned; no significant operating difficulties were encountered. The process is controlled via in-line pH measurements for the acid-deficient uranyl nitrate solutions. Information was developed on pH values for uranyl nitrate solution vs NO_3^-/U mole ratios, resin loading kinetics, resin drying requirements, and other resin loading process parameters. An engineering-scale resin loading

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system is being operated to develop and demonstrate all the process operations (Fig. 1) at the recycle pilot-plant scale. The feed resin is pretreated to control the size, shape, and purity of the product. The uranium leaves as dried resin ready for carbonization, the nitrate as $NaNO_3$ or NH_4NO_4 waste, and the water as condensate thus giving an efficient utilization of the uranyl nitrate feed solution.

Calculations made to estimate the capacities of equipment which is geometrically safe with respect to control of nuclear criticality indicate 100 kg/day or more of uranium for a single nitrate extraction line with four batch loading contactors. The four batch contactors might be replaced by a single continuous resin loading contactor of a modified Higgins contactor design. The resin loading process using solvent extraction of nitrate appears practical for use in a commercial-scale HTGR fuel recycle plant.

1. Paul A. Haas, "Loading a Cation Exchange Resin with Uranyl Ions," U.S. Patent 3,800,023, March 26, 1974.

2. P. A. Haas, "HTGR Fuel Development: Loading of Uranium on Carboxylic-Acid Cation Exchange Resins Using Solvent Extraction of Nitrate," ORNL-TM-4955 (September 1975).

3. C. C. Haws et al., "Engineering-Scale Demonstration of the Sol-Gel Process: Preparation of 100 kg of ThO_2-UO_2 Microspheres at the Rate of 10 kg/day," ORNL-4544 (May 1971).

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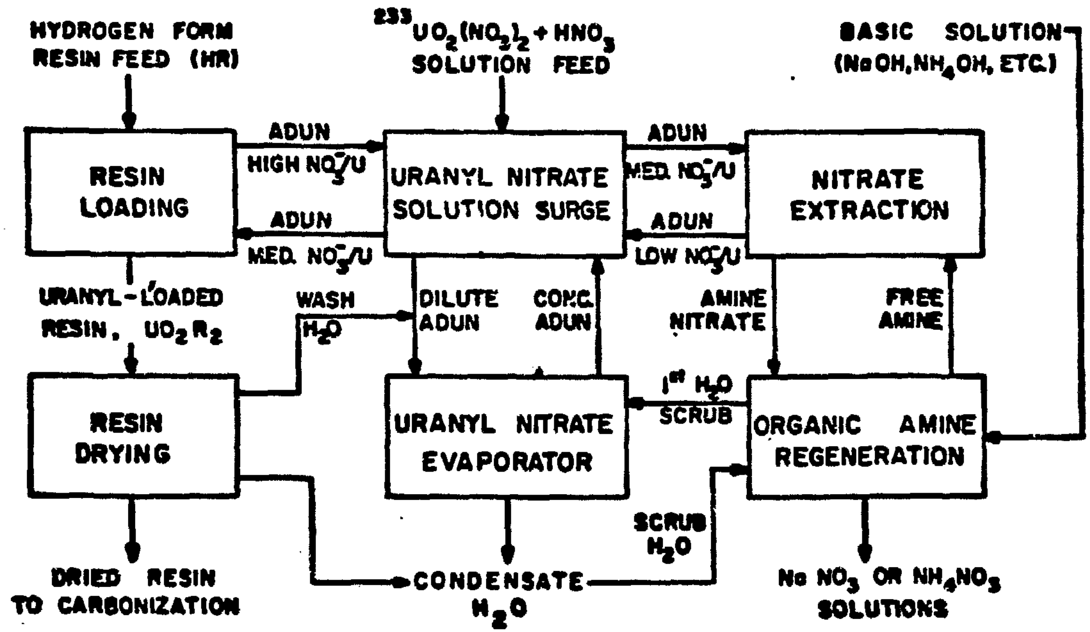

"ADUN" INDICATES ACID-DEFICIENT URANYL NITRATE ($NO_3/U<2$, MOLES/MOLE)

Figure 1. Schematic Reference Flowsheet for FRPP Resin Loading.