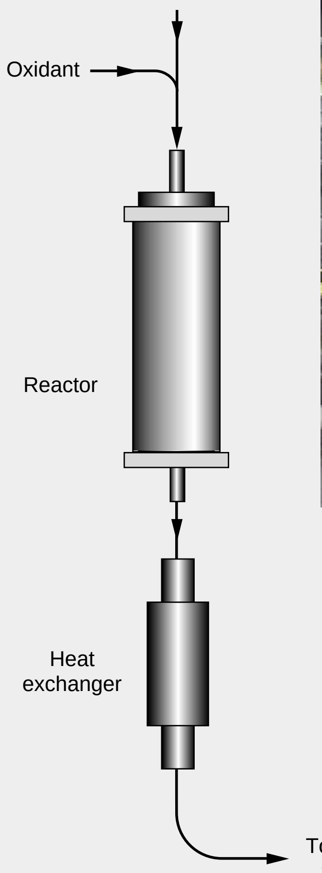
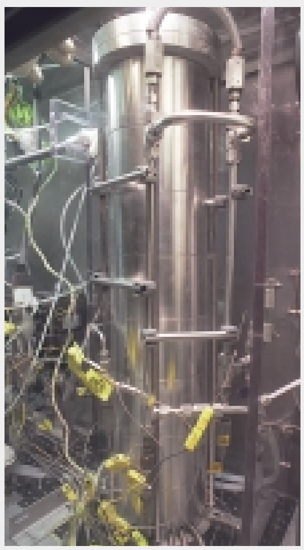
Hydrothermal Processing

Laura A.Worl, Steven J. Buelow, and Dennis D. Padilla





The reactor inside a glove box

Hydrothermal processing (reactions in hot water) offers the ability to destroy radioactive combustible wastes and is a viable alternative to incineration or costly storage options. For aqueous/organic mixtures, pure organic liquids, or contaminated combustible solids such as ion exchange resins, plastics, and rags, hydrothermal processing destroys more than 99.9 percent of the organic and nitrate components and facilitates the collection and separation of the actinides. The volume of TRU and mixed waste can be reduced by a factor of 100. The actual volume reduction depends on the type of waste being destroyed.

A schematic of a laboratory-scale process unit is shown in the Figure. The combustible waste is mixed with an oxidant (oxygen, air, or hydrogen peroxide) in water at pressures and temperatures above the critical point of water (374°C and 22.1 MPa). Under these conditions, water is a fluid with density high enough that reasonable process throughput can be achieved, but with transport properties like those of a gas, allowing rapid chemical reactions. Supercritical water is a unique solvent medium in which oxidation can take place at lower temperatures than those of incineration, thus limiting the production of NO_x and ash. Organic components are essentially completely oxidized to carbon dioxide and water by reaction with the supercritical water and oxidant. Chlorine-, sulfur-, and phosphorus-containing constituents are oxidized and converted to acids or salts, depending on the final pH of the product solution. At temperatures above 500°C, reactions are rapid and greater than 99 percent conversion is achieved in seconds.

The reaction is carried out entirely in an enclosed pressure vessel (the reactor) containing dilute reactants, so that the heat of reaction is absorbed by the solvent. The temperature within the reactor can be maintained at any desired level, typically in the range of 400 to 650°C. After about one minute of reaction, the mixture is cooled in a heat exchanger and depressurized. Any insoluble particles can be readily separated by a simple filtration step from the aqueous effluent. The effluent itself is further treated by polymer filtration for reuse or discharged as industrial waste.

We have conducted several experiment series using different waste matrices. In one series, actinide-contaminated organic mixtures reacted in 30 wt % H_2O_2 . The total organic concentrations at the inlet of the pressure vessel were between 1800 and 25000 mg/L, depending on the organic mixture. Under these conditions, the residual organic content was below 20 mg/L and in most cases below 5 mg/L. From these tests, the organic destruction efficiencies were greater than 99.9%. Experiments also were conducted with ion-exchange resins, including Reillex HPQ resin. For these experiments, the resin was mixed with water and a small amount of sodium salt of carboxymethyl cellulose, which is a viscosity enhancing agent that improves the pumping characteristics of the mixtures. The viscosity of the mixture is high and the resin beads do not settle out in a 24-hour period. After one minute, the reaction produced carbon dioxide, water, nitrogen, and hydrochloric acid. The levels of the total organic carbon were reduced to near our detection limits of about 6 mg/L.

A "hot" unit and a "cold" pilot unit have provided operational data for a full-scale unit that is expected to be in operation in 2001.

Hydrothermal processing will decompose plutonium-contaminated organic waste, such as contained in the bottle, into simple organic products such as CO_2 and water. Any insoluble actinide-containing material is removed by filtration, while actinides in solution are removed by polymer filtration.