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NUCLEAR MATERIALS AND FUEL CYCLE SERVICES SOURCES, INVENTORIES AND STOCKPILES

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heavy hydrogen or deuterium is increased by successively raising and lowering the temperature difference between water and a gaseous hydrogen compound (H_2S). All naturally occuring hydrogen compounds contain some deuterium which can be extracted. The process is based on the fact that deuterium migrates to the water stream at low temperature and to the hydrogen sulphide gas at high temperature. By suitable arrangement of the flow in separating towers, deuterium can be extracted from a feed of ordinary water. In each tower the water flows down through a series of perforated plates, while the hydrogen sulphide bubbles up through the traps. This arrangement promotes efficient mixing.

In this manner, the hydrogen gas is enriched in deuterium and, leaving the top of the hot towers, passes into a cold tower where deuterium migrates to the water feed. A portion of the H_2S gas stream enriched in deuterium is extracted and passes to the next stage. This process is then repeated in the second stage and in a further stage. Enriched water from the third stage then passes to a finishing section where it is distilled to a reactor product that is 99.75% pure D_2O.

12. Reprocessing

The earliest known reprocessing occurred at the Hanford Laboratories in the early-to-mid 1940's. The process utilized bismuth phosphates and lanthanum fluorides to precipitate plutonium from the low burn-up, metallic fuel in a batch process (since the sole objective was to obtain plutonium for weapons, residual uranium was discharged with the fission product wastes).

Apparently, the process worked well, achieving acceptably high efficiencies for fission product removal and plutonium extraction. However, the waste volume was high.

Other than the very fact that it worked, the primary lasting technological achievements of the Hanford facility were the concepts of remote operation and maintenance.

The next technological steps took place in both the US and the UK in the late 1940's and early 1950's. In both countries, the objectives were to develop a process which could operate continuously and which would recover uranium as well as plutonium.

At Hanford, the basic solvent extraction process was refined and accepted. In this process, countercurrent flows of aqueous and organic solutions are made to move through some sort of mixing chamber (column, bowl, tank, etc.). The organic solvent strips both uranium and plutonium from the aqueous feed solution, leaving the fission products behind. Later, by adjusting valence, the plutonium can be made insoluble, thus separating the uranium and plutonium.

Hanford called this the Redox process and used Hexone (Methyl-isobutyl Getone) as the organic solvent. While the process indeed achieved the objectives of continuous operation and extraction of both uranium and plutonium, Hexone was expensive and flammable and the process generated very large quantities of waste.

The UK, constructing a separations plant at Windscale, developed a similar process but utilized a different solvent - Butex. Its major advantage over the US solvent, Hexone, was a significant reduction in waste volume.

Finally, during construction of the Savannah River Plant near Aiken, South Carolina in the early 1950's, the Purex process was developed. This process was tributylphosphate (TBP) dissolved in a kerosene-like solvent. TBP has advantages of being chemically stable in nitric acid, relatively cheap, generating low waste volumes, and superior separations capability.

The basic Purex process is now generally accepted as the fundamental operating process for all reprocessing plants (with the notable exception of the abortive GE Morris effort). Different head ends (the means of dissolving the fuel and chemically preparing it for entry to the Purex process) and different final conversions (the means of converting the recovered uranium and plutonium to the desired chemical/physical form) are used depending upon the type of fuel to be reprocessed and the end-use of the recovered products.

Virtually all development work from then on has focused upon the methods used to bring the aqueous and organic liquid streams together and then separate them. The apparatus to do this is called a contactor. This is, of course, the heart of the solvent-extraction process and the key to both the process efficiency and waste volume.

Simple vertical columns were used as the first and most obvious contactor. These extraction columns were packed with various metal or ceramic shapes to create a very long flow path. The heavy aqueous solution was introduced at the top of the column and allowed to flow downward under the influence of gravity. The lighter organic solution, introduced near the bottom of the column, was displaced by the aqueous solution and forced to flow upwards. Thus, a countercurrent flow could be established and because of the metal or ceramic shapes, intimate aqueous/organic contact occurred. However, since flow rates were low and aqueous/organic contact was gentle, the columns had to be very tall to achieve reasonable efficiency. (The process building at Windscale is said to be 20 stories tall.)

The first step in the evolution of more efficient contactors involved pulsed columns. Pulsed columns utilize multiple perforated plates and, by applying alternating positive/negative pressure "pulses", force the liquids to pass back and forth through the perforations. This imparts a vigorous mixing action of the two streams. Thus, even though the two liquid streams still moved only by gravity, the extraction efficiency was significantly improved and column height could be reduced. This type of pulsed extraction column was used at the Hanford, Idaho National Engineering Laboratory (for naval propulsion and research reactor fuel reprocessing), and Nuclear Fuel Services' plants.

The next evolution in contactors involved a device called a "mixer-settler". This was a horizontal device (i.e. - required much lower buildings) of multiple stages. In each stage, the aqueous and organic streams are first drawn together and vigorously "mixed" by an agitator. Then the mixed solutions are driven by the agitator into a long, horizontal "settling" chamber. In this chamber, gravity again takes effect and the lighter organic solvent rises to the top while the heavier aqueous solution settles to the bottom. Both solutions are then separately drawn off from the chamber and introduced to further mix/settle stages.

This contactor must be considered a major step in that the mixing action was very strong and - for the first time - the two streams were mechanically propelled through the stages. Since the device was horizontal it was very amenable to either remote maintenance (as in its first use at Savannah River) or to a design where the mixer motors were physically remote from the mixers (as at later Windscale facilities). Unfortunately, the design inherently involves large volumes of mixed solutions and thus, major inventories of dissolved uranium and plutonium. This in turn causes considerable chemical and radiolytic solvent degradation - involving both solvent makeup expense and somewhat increased waste volumes. This problem was the driving force to further improve contactor performance.

The centrifugal contactor, developed at Savannah River, was the next improvement. In this device, the mixed solutions are forced to flow from the motor driven agitator to a small centrifugal separator bowl mounted on the same shaft. In this bowl (which effectively replaces the long horizontal settler chamber), the aqueous and organic solutions are separated by centrifugal force.

In this device, for the first time, mechanical force was applied to all three primary solvent extraction sub-processes - stream movement, stream mixing, and stream separation. The results are impressive, including much lower in-process inventory (~25% of a mixer-settler), high separations efficiency, and low solvent degradation (and therefore, lower waste volume). Because of these advantages, Savannah River replaced their existing mixer-settler contactors with centrifugal settlers.

A derivative of the centrifugal contactor is the multi-stage centrifugal contactor developed by Saint-Gobain Techniques Nouvelles. This contactor, called Robatel, incorporates the equivalent of eight separate centrifugal contactors on a single motor driven shaft. This device is used in the first extraction stage at the AGNS Barnwell facility. Presumably, it may also be used in the planned new French plants and in any plants exported by the French.

Fuel reprocessing technology has been well established over the last twenty years in several countries, particularly for low burnup metallic fuels. Large scale experience has not yet been obtained in handling high-burnup oxide fuels.