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FLUIDIZED BED VOLATILITY -- PROCESS DESIGN FOR BrF_5 FLUORINATION AND UF_6 RECOVERY

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ABSTRACT

A process design is presented in which a single piece of equipment is used for (1) UF_6, BrF_3 and BrF_5 recovery; (2) UF_6 purification; and (3) conversion of BrF_3 to BrF_5.

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INTRODUCTION

BrF_5 is being considered as an alternate fluorinating agent in the fluid bed volatility process because it has the following advantages over F_2: (1) lower heat of reaction with U_3O_8 (easier temperature control in the bed) and (2) BrF_5 will convert UF_4 to UF_6 but will not convert PuF_4 to PuF_6 thereby effecting a U-Pu separation in the fluorinator (the Pu is subsequently recovered as PuF_6 using F_2 as the fluorinating agent). Since unreacted BrF_5 and the reaction product BrF_3 must be condensed with the UF_6 and separated, the use of BrF_5 adds some complexities to the flowsheet. Previous flowsheets have included (1) cold traps for BrF_5, UF_6, BrF_3 collection, (2) a distillation column (or columns) for UF_6 separation, and (3) equipment for conversion of BrF_3 back to BrF_5. Since the process, as presently envisioned is batch, these flowsheets involve considerable surge tankage between steps. An improved flowsheet has been developed which incorporates a single system for BrF_3-BrF_5-UF_6 collection, UF_6 separation, and BrF_3 fluorination and allows recycle of unreacted BrF_5 and conversion of BrF_3 to BrF_5 during the fluorination step.

Description of the System

A schematic diagram of the flowsheet is shown in Fig. 1. As in the conventional flowsheet the initial step is decladding the fuel with either HF-O_2 or HCl. During or after decladding the UO_2 is oxidized to U_3O_8 and then converted to UF_6 by the following overall reaction:

$$9BrF_5 + U_3O_8 \rightarrow 9BrF_3 + 3UF_6 + 4O_2$$

During this conversion the equipment operates as follows: BrF_5 vapor (plus inert gas if necessary to maintain fluidization of the bed) is introduced into the bottom of the fluid bed reactor. The gases leaving the top of the fluid bed reactor, consisting of UF_6, BrF_3, unreacted BrF_5 and O_2 (+ inert), are introduced into the bottom of the scrubber where UF_6 (B.P. $\sim 64°C$) and BrF_3 (B.P. 125.7°C) are scrubbed from the inerts by refluxing BrF_5 liquid (B.P. 41.3°C). The BrF_5, in excess of

that required for holdup, leaves the tower as a vapor from the reflux drum and is condensed and recycled to the fluid bed reactor via a surge tank, pump and vaporizer. The inerts, containing some low boiling F.P.'s are sent to cleanup before venting. During this cycle the UF_6-BrF_3 solution accumulates in the reboiler of the scrubber. F_2 is introduced into the scrubber at this time, if desired, to convert some of the BrF_3 to BrF_5 for recycle to the fluid bed reactor. A heel of BrF_3 must be maintained, however, to act as a reboiler liquid and "carrier" for the UF_6. After all of the U_3O_8 has been removed from the fluid bed, the scrubber system is isolated from the fluid bed reactor and the Pu is removed from the bed by conversion to PuF_6 using fluorine. The PuF_6 is recovered in a separate system and the F_2 recycled.

During the fluorination of the plutonium, the scrubber is operated as a batch distillation column for separation and recovery of BrF_5, UF_6 as follows:

> The column pressure is raised to above the triple point of UF_6 (1.5 atm) and the BrF_5 holdup in the column is distilled overhead. Some UF_6 will be included in this cut and recycled to the next fluorination in order to provide a pure UF_6 product. Next, the UF_6 is distilled overhead and drained to a product tank. Again to provide a pure UF_6 product, a small cut of UF_6-BrF_3 must be taken overhead and to the BrF_5 recycle tank for recovery in the next cycle. The fission products which accumulate in the recycle system are removed by periodically purging some BrF_5 (during the part of the cycle before UF_6 is distilled from the column) and BrF_3 (after all UF_6 has been distilled from the column).

Advantages of the System

Besides the obvious advantage of fewer pieces of equipment and simplicity, this flowsheet has the additional advantages that (1) a more efficient collection system, liquid scrubbing, is used for recovery of UF_6 (low temperatures are not required for high UF_6 recovery);

(2) unreacted BrF_5 can be recycled to the fluid bed reactor; (3) BrF_3 can be continuously converted to BrF_5 and recycled during the fluorination, thereby reducing BrF_3 + BrF_5 holdup requirements; and (4) the BrF_5 reflux takes care of the BrF_3 + F_2 heat of reaction.

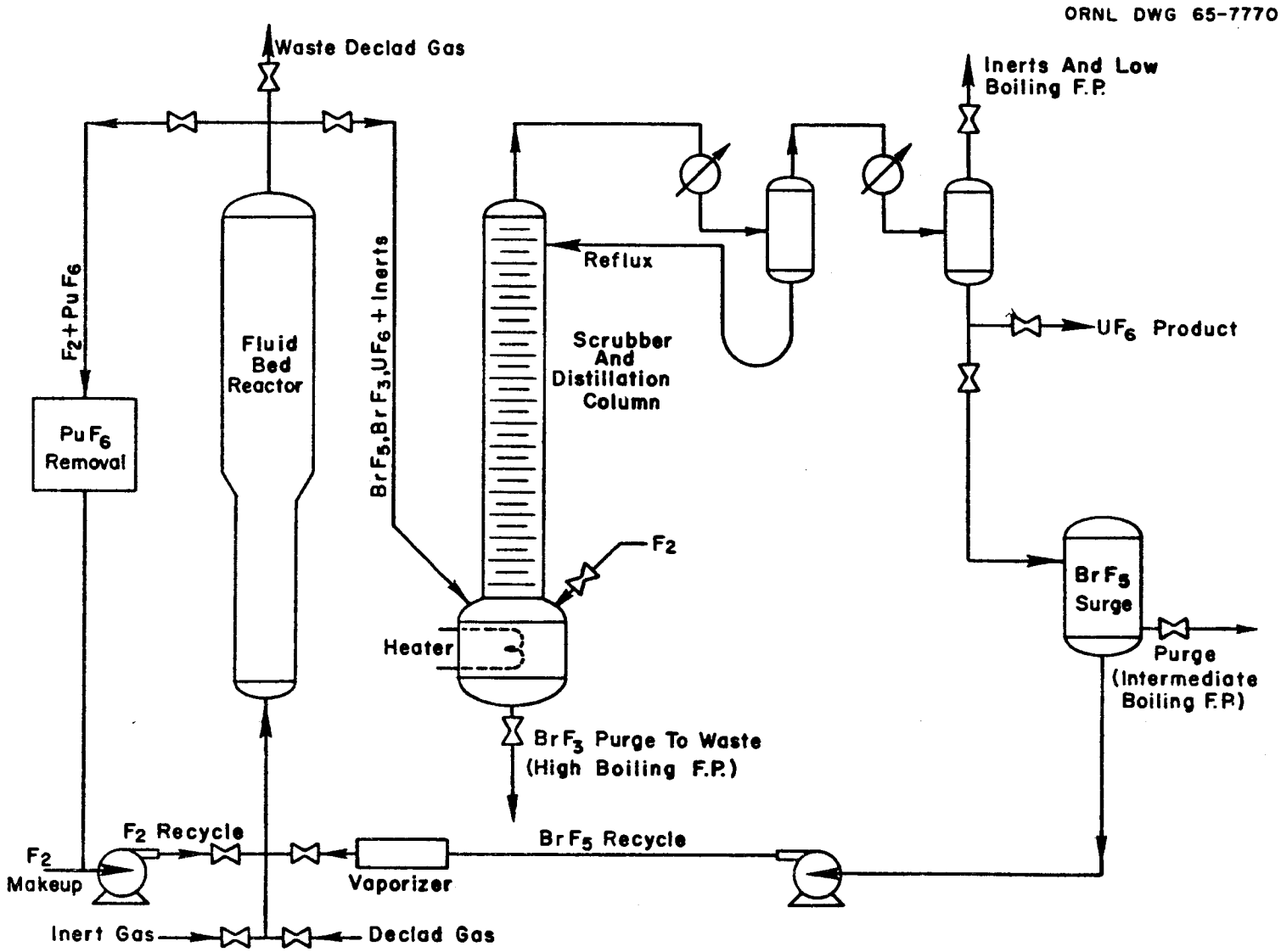

Fig. 1. BrF_5 Fluid Bed Volatility Flowsheet.

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