

Recycling hydrofluoric acid in the nuclear industry: The OverAzeotropic Flash process (OVAF)



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ARTICLE INFO

Article history:

Received 11 June 2015

Received in revised form 19 October 2015

Accepted 3 March 2016

Available online 10 March 2016

Keywords:

Hydrofluoric acid

Recycling

Anhydrous fluorine gas

Nuclear fuel cycle

ABSTRACT

One of the greatest uses for fluorine is in the nuclear industry, to make uranium hexafluoride, needed to produce the U-235 isotope. Fluorine could in theory be recycled back to produce the hexafluoride as fluorine free uranium is needed for fuel manufacture but previous efforts to recover fluorine were unsuccessful because either a too large energetic cost, safety and or corrosion issues. Potential modifications of the current process to recover fluorine are analyzed, on the basis of partial reactions involved in the deconversion process, leading to a global solution based on the use of an extra overazeotropic flash drum to the existing industrial process.

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1. Introduction

Uranium and fluorine are intimately bound in the nuclear fuel cycle. Natural uranium has an insufficient isotopic 235 content to be used in the current light water nuclear reactors. Therefore, it has to be enriched in 235 isotope by separating it from the main 238 isotope in an efficient way. The only two processes, which have nowadays reached industrial large scale, namely gaseous diffusion and ultracentrifugation, require the use of uranium in gaseous state. Fluorine is the only chemical element that added to uranium, allows the generation of a stable gaseous molecule, the uranium hexafluoride (UF₆) at moderate temperature and benefits also from the advantage of a single stable isotope.

Depleted uranium can be deconverted in oxide form and can be used again, either to prepare MOx fuel combined with plutonium or stored for future use in fast breed reactors. On the other hand, fluorine is currently not recycled in the U cycle. To be efficiently recycled in the U cycle, it has to be preferably produced as anhydrous fluoride gas (AHF), as raw material for either uranium oxide fluorination into uranium tetrafluoride UF₄ or fluorine electrolysis [1] (Fig. 1).

HF is produced in a hydrated form in the current industrial process. The presence of an azeotrope around 38% weight HF (Fig. 2) makes this dehydration problematic and has not been yet

implanted. Therefore, fluorine is either sold in aqueous form for external use or stored without deconversion in UF₆ form. This storage presents environmental, health, and safety risk because of the UF₆ chemical instability: in contact with water or moisture chemical reactions producing hazardous material such as HF can occur.

The main alternatives to the current industrial deconversion process to produce anhydrous fluoride gas have been discussed by Morel [1]. Most of them use a third body extraction, which is problematic to conserve the uranium and fluorine compounds purity. Morel concluded that probably the best way was to use the chemical reactors of the current industrial process to remove the extra water. However, the different attempts [2–4], faced either corrosion and flow rate issues or too high energetic cost.

In this paper, the current industrial process is analyzed with thermodynamic and kinetic data. Modifications of this process producing AHF but with lower energy extra-cost, along with limited corrosion and safety issues will be discussed.

2. Baseline of the current industrial process

The current industrial process is split into two phases (Fig. 3): a hydrolysis of UF₆ into UO₂F₂ and a pyrolysis of UO₂F₂ into either U₃O₈ (for storage) or UO₂ (for fuel application). The hydrolysis of UF₆ is done at medium temperature (200–300 °C) in gas phase and the pyrolysis is done at high temperature (650–800 °C) with a mixture of hydrogen and water. The exhaust gas streams of pyrolysis are injected in the hydrolysis section and the exhaust gas

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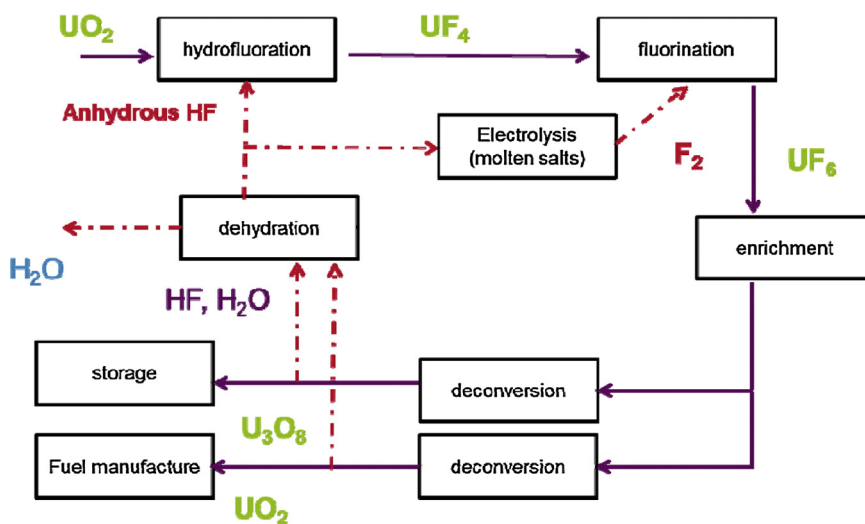


Fig. 1. French nuclear fuel cycle and recommended pathway to recycle fluorine.

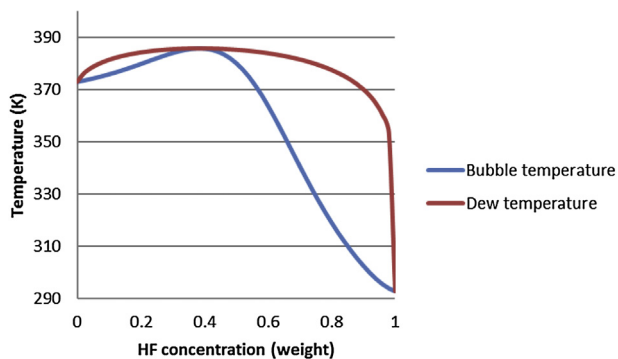


Fig. 2. HF-H₂O mixture vapor-liquid equilibrium at atmospheric pressure.

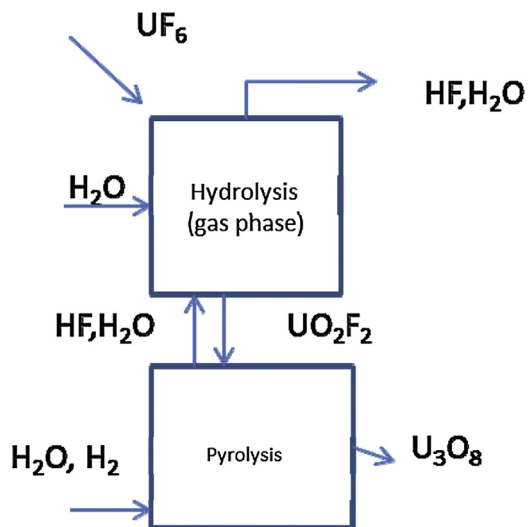


Fig. 3. Schematic of the current industrial process.

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