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Thermodynamic analysis of fluorine regeneration from uranium hexafluoride with a flame of hydrogen-containing fuel and oxygen-containing oxidant using

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The uranium hexafluoride depleted in the U-235 isotope (DUHF) is a by-product of isotope uranium enrichment in the nuclear fuel cycle (NFC). The accumulated DUHF amount in the world reaches 2 million tons, including in Russia up to 1 million tons.
Uranium hexafluoride is a very toxic substance (maximum allowable concentration in the air is 0.015 mg•m-3), volatile (p = 0.1 MPa at 56 ℃) and prone to hydrolysis. Therefore, DUHF large quantities storing is a risk to the environment.
At the same time in the composition of DUHF there is a fluorine, the important chemical substance that is advisable to regenerate.
The only industrial technology of DUHF processing developed by the "AREVA":
UF6 gas + 2H2Ogas ⟶ UO2F2 sol + 4HFgas, T = 250-400 oC; (1)
3UO2F2 sol + 3H2Ogas + H2 gas ⟶ U3O8 sol + 6HFgas + H2Ogas, T = 450-600 oC. (2)
At this temperature the most stable uranium compound is UF4. In order to avoid UF4 formation and to regenerate fluorine completely, process (2) is carried out with an excess of water. Therefore, the product of this process is an azeotropic aqueous solution of HF, which does not allow the NFC closing for fluorine, because the uranium fluorides production based on anhydrous hydrogen fluoride.
The authors performed a thermodynamic analysis of the possibility of HF obtaining from the UF6 in the flame of the hydrogen-containing fuel and oxygen containing oxidant:
UF6 gas + CmHnNk gas + O2 gas ⟶ UOx sol + mCOy gas + 6HFraз + 0.5kN2 gas –Q, (3)
where CmHnNk –hydrogen-containing substance, m≥0, n>0, k≥0, x=2÷3, y=1 or 2.
For elements U-H-O-F-N the thermodynamically equilibrium composition of substances were calculated depending on temperature and the ratio of atoms. The calculation was developed based on the method of minimization of Gibbs energy on the molecular composition. It was shown that at temperature above 1100 K at a specific ratio of atoms hydrogen fluoride is practically the only fluorine-containing substance, and uranium dioxide is virtually the only uranium-containing substance.
This temperature in the reaction zone can be obtained using the process of recovery of UF6 in the combustion regime (3). For hydrogen, methane, ammonia as a fuels and oxygen and air as an oxidizers were calculated adiabatic temperature of the products and temperature taking into account thermal radiation of a heterogeneous flame. The values of these temperatures for the stoichiometric ratio of the HF obtaining in all cases exceeded 1100 K.
Also, we calculated the cooling rate of the products of the process necessary to minimize the rate of reverse reaction of uranium dioxide fluorination with hydrogen fluoride.
Thus, the recovery of UF6 in the flame of the hydrogen-containing fuel and oxygen-containing oxidizers at temperatures above 1100 K, and fast cooling products can become the basis for a method of hydrogen fluoride obtaining from DUHF.

Primary authors: Prof. PASHKEVICH, Dmitrii (Peter the Greate Saint-Petersburg Polytechnic University; New Chemical Products Ltd.); Mr ZIMIN, Arsenii (New Chemical Products Ltd.)

Presenter: Prof. PASHKEVICH, Dmitrii (Peter the Greate Saint-Petersburg Polytechnic University; New Chemical Products Ltd.)

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