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Ammonium Nitrate and Chelating Agents Decomposition in Autoclave During ILW Processing

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A large amount of intermediate level waste (ILW) containing ammonium nitrate, as well as some chelating agents and precipitator, such as diethylene triamine pentaacetic acid (DTPA) is generated during spent nuclear fuel reprocessing which can not be included into vitrified HLW due to the presence of explosive components. ILW processing is proposed which includes ammonium nitrate significant decomposition using formalin during ILW evaporation with circulation of bottom product followed by autoclave treatment of the latter. The process was elaborated and tested with the use of the laboratory-scale automated rigs.

Autoclave decomposition of organic sorbents by HNO3 [1] has been a base of the proposed procedure. The process of NH4NO3 decomposition in a batch mode with 30-50% loading of auto-clave by volume with the stimulant containing 2-6 mol/dm3 NH4NO3 and 2-6 mol/dm3 HNO3 is car-ried out for 5-6 h with an 98% efficiency at 180-210 ℃ generating vapour and gas pressure of 4.0-8.0 MPa. The main gaseous products are CO2 and N2O. The process could be performed at other component concentrations, depending on the conditions of the other reprocessing stages, particular-ly ILW evaporation, mentioned above.

DTPA degradation in autoclave is performed in 2 steps, the temperature of first and second stages are 130 oC and 210 oC respectively. This approach allowed us to reduce the sharp increase in pressure in the process and completely decompose both DTPA and the intermediate products of its decomposition.

The joint decomposition of NH4NO3 and DTPA in a two-step process has been also per-formed [2], indicating on decomposition of > 95% NH4NO3 and > 98% DTPA, respectively. How-ever, the working pressure during the process of joint ammonium nitrate and DTPA decomposition significantly exceed those of their individual decomposition and apparently is summed. The loading of the autoclave should be decreased to reduce the operating pressure in it.

The preliminary NH4NO3 decomposition by formalin during the evaporation is expedient-ly carried out at high NH4NO3 accumulation in ILW for its processing. The volume reduction factor during the testing of ILW stimulant evaporation has been [2]. Destruction efficiency has amounted to 91-93% at a molar consumption ratio CH2O:NH4NO3 = 2, while an amount of decomposed HNO3 has been threefold greater. These characteristics should be reduced to the level of the similar batch process [3], i.e. CH2O:NH4NO3 ≤ 1 without significant HNO3 decomposition.

Verification of the flowsheet as a whole confirmed that the above process characteristics are sufficient for commercial implementation.

1. Bagerman M.R., Onufrienko C.V., et al. Patent RU № 2062517, Bul. 17 (1996).
2. Mishina N.E., Murzin A.A., et al. Patent application RU № 2013130551 (02.07.2013)
3. Bartenev S.A., Zilberman B.Ya., et al. Patent RU № 2329554. Bul. 20 (2008).

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