Handling of Ammonium Nitrate Mother-Liquid Radiochemical Production – 13089

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ABSTRACT

The aim of the work is to develop a basic technology of decomposition of ammonium nitrate stock solutions produced in radiochemical enterprises engaged in the reprocessing of irradiated nuclear fuel and fabrication of fresh fuel.

It was necessary to work out how to conduct a one-step thermal decomposition of ammonium nitrate, select and test the catalysts for this process and to prepare proposals for recycling condensation.

Necessary accessories were added to a laboratory equipment installation decomposition of ammonium nitrate. It is tested several types of reducing agents and two types of catalyst to neutralize the nitrogen oxides.

It is conducted testing of modes of the process to produce condensation, suitable for use in the conversion of a new technological scheme of production.

It is studied the structure of the catalysts before and after their use in a laboratory setting.

It is tested the selected catalyst in the optimal range for 48 hours of continuous operation.

INTRADUCTION

In the technology of reprocessing of irradiated nuclear fuel and the production of fresh oxide UO_2 fuel one of the species produced in large quantities LLW is the mother liquors from the operation of chemical denitrification. This operation is performed by uranium precipitation with ammonium hydroxide to form a precipitate poliuranata ammonium. The main component of salt-containing mother liquor is ammonium nitrate. This type of LLW can not be disposed of in the form of direct cement compound. To address the issue of a possible burial stock solutions must first remove them from ammonium nitrate.

To this end, JSC "VNIINM" together with JSC "SCC" (Siberian Chemical Combine) was developed and tested method of thermal decomposition of ammonium nitrate in the reactor with ohmic heating.

The method consists in the following. Initial stock solution is mixed with a reducing agent and fed to the reactor. The reactor solution is sprayed by the carrier gas and under the influence of high temperature (500 °C), ammonium nitrate decomposes. A large proportion of the nitrogen oxide reacts with a reducing agent to form water, nitrogen and carbon dioxide. Inside the reactor is resistant to chemicals and mechanical valve (or) ceramic catalyst. Tests have shown that this method can achieve the degree of decomposition of ammonium nitrate, more than 99.9%. The condensate, depending on the conditions of decomposition contains either dilute nitric acid or ammonium hydroxide.

The described method of disposal of ammonium nitrate stock solution is optimal for the following reasons:

- the degree of decomposition of ammonium nitrate in a single step up more than 99.9%;
- not form secondary LLW;
- all non-degradable components of the mother liquor concentrated on mechanical support and further disposed as solid waste. Concentration under this reaches more than 10^6;
- resulting acidic condensate may be used within technological scheme.

To implement the method of thermal destruction of ammonium nitrate queen cells need to pick up a steady catalyst to determine the optimal mode of installation. Resolution of these issues is the aim of the present work.

Main part

To do the work a laboratory apparatus was used and shown in Figure 1.

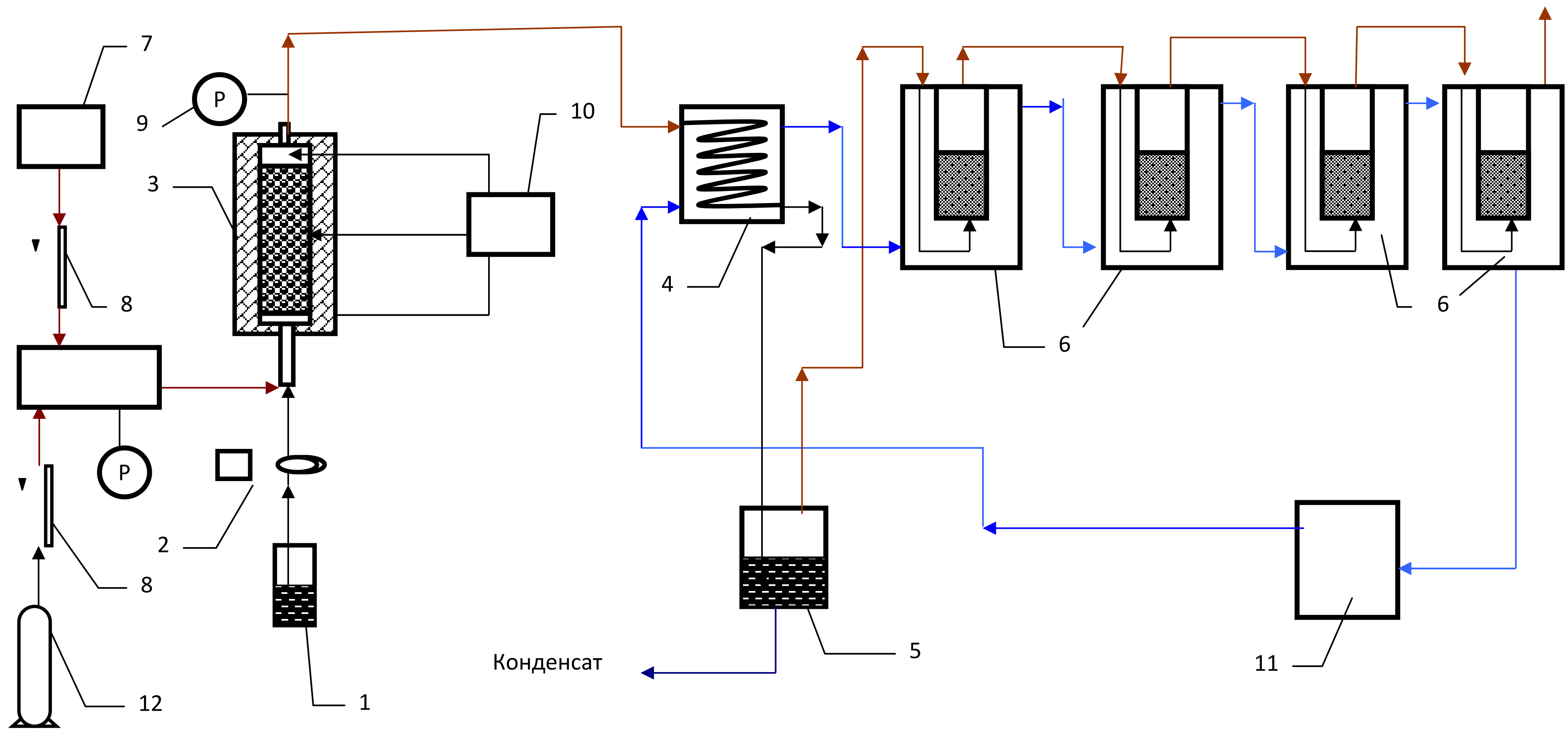
The initial solution from the tank 1 is pumped into the bottom of two capsules, where it is sprayed with gas supplied from a cylinder 12 and the blower 7. Dispersed solution is fed into the reactor expansions 3. The capsule, filled with packing and catalyst of the decomposition of components supplied to the solution, and then steam-gas mixture enters the refrigerator 4, where the water vapor condenses and enter the collection 5. Collection 5 also serves as a separator and separation of gas from the liquid. The gas then enters the series four similar adsorbing column 6, where the absorption of various reagents gas components of the reaction products. Next, the gas stream is discharged to the atmosphere of boxing.

For the condensation of water vapor in the external circuit is fed water cooler 4 of hladostata 11. For a more complete absorption of water in the gaseous products of the reaction temperature is maintained columns 6 (5 ± 0,5) °C by hladostata 11.

Operating temperature reactor 3 set with control unit 10. During the process of decomposition of ammonium nitrate, and the temperature is controlled at three points in the reactor 3. For each heater was set their temperature.

During the experiment at regular intervals the condensate was drained from the tank 5. In analyzing the value of the condensate pH, the concentration of nitrate ions, the concentration of free ammonia content of dry residue.

To determine the composition of the gas phase during the experiment leaking (the condenser) a fixed time, and then samples were taken from the absorption column.



1- Capacity of the original solution; *2*- pump; *3*- decomposition reactor; *4*- refrigerator; *5*- condensate collector; *6*- Absorption column; *7*- blower, *8*- rotameter; *9*- manometer; *10*- control block; *11*- Hladostat; *12* – Nitrogen bottle

Picture 1 Scheme of the laboratory setup for the decomposition of nitrate-ammonia solutions.

2 Results and Discussion

In all experiments, the model solution simulating the composition of the mother liquor of the following composition [NH_4NO_3] = 2,75 mol / l, [NH_4OH] = 0,05 mol / l, [U] = 10 mg / L, [Fe] = 2 mg / L, pH = 7.92. To this solution was poured in the required number of solutions of reducing agents. Chosen as the reducing formaldehyde, urea and ammonium hydroxide. As stock solutions of reducing agents used 37% of the masses. Formaldehyde, 25% by weight. ammonia and 7.6 mol / urea.

Temperature range coating area and decomposition of ammonium nitrate in all experiments was 600 °C at the heater. In the area of reduction of nitrogen oxides (middle and upper heater) temperature was maintained at 550 °C. Without filing solution temperature difference between the wall heater and central part of the capsule was 100 °C. After submission of the solution in the bottom of the temperature difference increased to 150-200 °C in the middle and upper parts within 110-120 °C.

Conclusion

The results show that the designed process of thermal degradation of ammonium nitrate queen cells can be recommended for industrial production.

In general, the work has the following conclusions

- Process of safety thermal decomposition of ammonium nitrate-queen cells - technical feasibility process, including a commercial scale;
- Used as a reducing agent - ammonia as the reagent is already being used in the circuit NPC and does not create additional problems with the cleaning of exhaust gases and the use of condensate stripping uranium;
- When the mother liquor spray air with any of the tested reductants not possible to get a neutral environment in the condensate and hence the complete absence of nitrogen oxides in the effluent gases;
- For condensate with a pH of 4.8 is necessary to conduct the process in oxygen-free environment, and maintaining relationships with the exact ammonia nitrogen to nitrate nitrogen in the range 1,2 ± 0,1 (molar ratio). To this end, the development of design documentation to work out the design of spray devices enabling after starting the switch to spray steam with low degree of overheating;
- To reduce the cost of the process may lead the expansion of the mother liquor in air, but the ratio of N (-3) / N (+5) should be raised to 1.4 to give a condensate with a pH of 1.4 to 1.7;
- To reduce the thermal degradation of ceramic catalyst is recommended to have it over a layer of spherical metal powder corrosion resistant in the gas phase;
- For the industrial installation required performance (2200 m3) unit should be made of several sections of the same type (tubular heaters) to ensure uniform heat transfer catalyst on length;
- For use in an installation recommend catalyst brand RK-205-04, as it showed the best results in physical strength, composition of flue gas condensation at medium acidity.