Complex utilization of snf processing wastes in air plasma of high-frequency torch discharge*

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Abstract. We present results of complex spent nuclear fuel wastes utilization process in air plasma of high-frequency torch discharge in form of dispersed water-organic compositions. We demonstrate the possibility to apply magnetic separation for effective extraction of obtained dispersed solid products including magnetic iron oxide from water suspension.

1. Introduction

The Russian Federation first in the world began to creating closed nuclear fuel cycle. It provides for delivery spent nuclear fuel (SNF) from NPP, its storage, recycling and extraction Pu-239 and U-238 to produce MOX fuel based on these elements and further delivery back to NPP. Closed nuclear cycle conception also provides for utilization all the nascent radioactive wastes, thus providing long time storage and further using of them.

Tadiochemical plants have SNF recycling technologies based on PUREX process. It provides high level extraction of uranium and plutonium (>99,9%) with high level purification of them from fission products [1-3].

SNF extraction technological schemes provide for application tributylphosphate (TBP) with different diluents (kerosene, refined hydrocarbons, carbon tetrachloride, hexachlorobutadiene (HCBD), etc.) as an extractant for uranium and plutonium extraction. For natural and low enriched uranium concentration of TBP is 30÷40%. In the case of highly processed U-235 fuel or fuel with high plutonium concentration TBP content is reduced to 2,5÷5% in order to avoid the formation of dangerous concentrations of fissile nuclides in the extract [2].

Under the influence of radiation exposure due to a high content in the solution of fission products, plutonium and transplutonium elements extractants over time lose their effectiveness and become inflammable recycling waste (IRW SNF).

After uranium and plutonium extraction from dissolved SNF there are also recycling wastes (RW SNF) are formed as low-concentration solutions with next modelling consist [4]: HNO_3 – 18,0%, H_2O – 81,43%, Fe – 0,07%, Mo – 0,1%, Nd – 0,11%, Y – 0,06%, Zr – 0,058%, Na – 0,04%, Ce – 0,039%, Cs – 0,036%, Co – 0,031%, Sr – 0,026%.

Next these wastes go to evaporation and after addition chemical reagents (silicates, phosphates, borates, etc.) go to verification with further landfilling [5]. Such technology is multistage and it requires significant energy, maintenance, chemicals and time.

In article [6,7] the processes of plasma utilization of SNF in the form of optimal inflammable water-organic compositions (WOC) on the basis of acetone and ethyl alcohol.

These WOCs have an adiabatic combustion temperature at least 1200°C and thus give the possibility of a significant reduction in energy consumption for the process of recycling.

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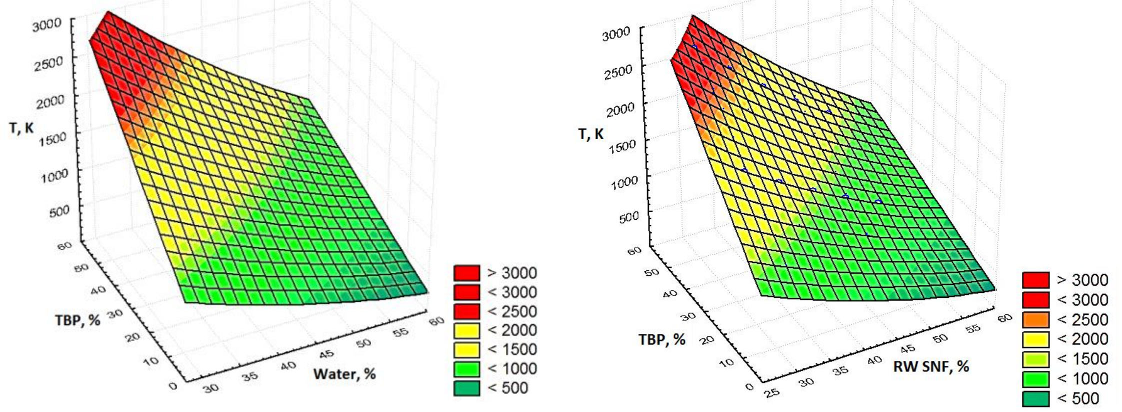
In this paper we consider the possibility of separate and joint IRW SNF and RW SNF plasma utilization as inflammable water-organic compositions.

2. Calculation of optimal consist of water-organic compositions

At the first stage calculations were made for burning indicators of different WOCs based on IRW SNF and RW SNF.

Figure 1 shows influence of content of TBP and water (a) and also TBP and RW SNF (b) on adiabatic combustion temperature (T) in water-organic compositions with different consists



a) b)

Figure 1. Influence of content of TBP and water (a) and also TBP and RW SNF (b) on adiabatic combustion temperature in water-organic compositions

Analyzing the results should be that replacement of water in inflammable WOC with low-concentration RW SNF solutions does not influence on T of WOCs. It points on possibility to utilize of IRW SNF and RW SNF joint in plasma.

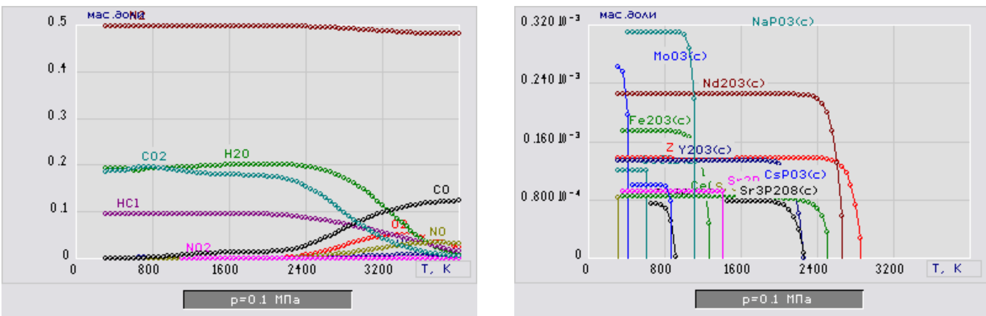
Considering all the findings the next optimal consists can be recommended for practical application:

- WOC-1 (50% water : 17,5% TBP : 32,5% HCBD);
- WOC-2 (50% RW SNF : 17,5% TBP : 32,5% HCBD).

Equilibrium consist calculations of gaseous and condensed plasma utilization products were done for recommended WOC-1 and WOC-2. These calculations were done by means of licensed program for PC called «TERRA».

3. Calculation of optimal plasma utilization regimes of water-organic compositions

Figure 2 shows characteristic equilibrium consists of the main gaseous (a) and condensed (b) products after joint plasma utilization of IRW SNF and RW SNF in for of WOC-2 with mass proportion of air plasma coolant 65%.



a) б)

Figure 2. Equilibrium consists of the main gaseous (a) and condensed (b) products after joint plasma utilization of IRW SNF and RW SNF in air plasma (65% air : 35% WOC-2)

Analyzing the results should be that with mass fraction of air coolant is 65% the main gaseous products (temperature under 1500 K) of RW SNF plasma utilization in form of WOC-2 are N_2, H_2O and CO_2.

At the temperatures up to 800 K the main condensed products are metal chlorides such as $FeCl_2$(c), $CoCl_2$(c) и $SrCl_2$(c) effectively attaching chlorine. In temperature interval 800-1500 K simple metal oxides form such as MoO_2(c), Nd_2O_3 (c), ZrO_2(c), Y_2O_3(c), CeO_2(c). Also in this interval complex phosphorus metal oxides form ($NaPO_3$(c), $CsPO_3$(c), $Sr_2P_2O_7$(c) и $Sr_3P_2O_8$(c)). They attach phosphorus effectively.

Small amount of soot C(c) and CO, NO, NO_2, HCl indicates that plasma utilization process of wastes in form of WOC-2 takes place in environmentally safe mode. It is worth noting formation iron oxide in condensed phase. Next increasing of air mass fraction to 90% reduces process productivity.

Thus joint plasma utilization of RW SNF and IRW SNF in form of water-organic composition with optimal consist eliminates evaporation stage, significantly reduces specific energy consumption on this process and also allows to use magnetic separation for effective extraction of particulate solid products of plasma utilization.

Table 1 shows calculated results of energy costs of the waste plasma utilization in form of optimal WOC-2 with with optimal air plasma coolant mass ratio (65%).

Table1. Energy costs of WOC-2 plasma utilization

T, K	500	1000	1500	2000	2500
Energy costs, MJ/kg	0,24	0,91	1,66	2,50	3,76

Considering all the findings the next optimal modes cam be recommended for practical application of plasma utilization process of SNF processing wastes in air plasma:

- WOC consist (50% RW SNF : 17,5% TBP : 32,5% HCBD);
- weight ratio of phases (65% air : 35% WOC);
- temperature interval (1200±100) K.

4. Experiments

Figure 3 shows scheme of plasma module based on high frequency torch (HFT) plasmatron. It is intended for high frequency air plasma generation and application of this plasma for carrying out different plasma-chemical processes.

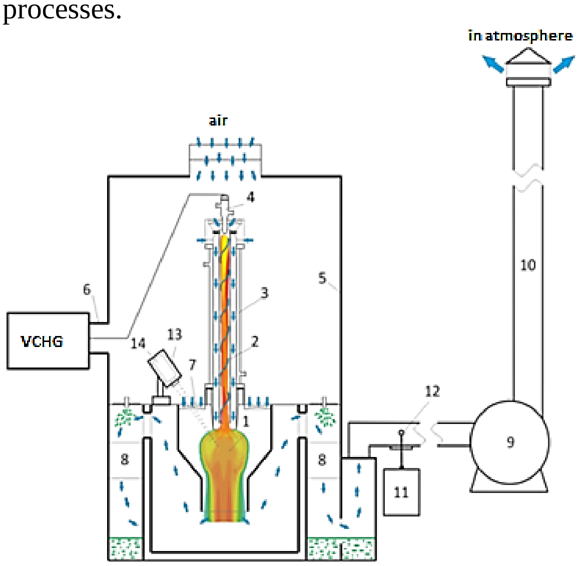


Figure 3. Scheme of plasma module based on generator VCHG8-60/13

1 – dispersant, 2 – HFT discharge, 3 – HFT plasmatron; 4 – copper electrode, 5 – case; 6 – coaxial output; 7 – reactor impeller; 8 – wet cleaning unit for exhausted gases; 9 – air exhauster (BP 12-26, №4), 10 – ductwork, 11 – gas analyzer «Quintox» КМ 9106, 12 – sampler; 13 – pyrometer protective cover, 14 – pyrometer IPE 140/45

Plasma module includes high frequency generator VCHG 8-60/13-01 (oscillatory power up to 60 kW, operating frequency of 13.56 MHz), from which via coaxial output 6 high frequency energy is supplied to the water cooled copper electrode 4 of high frequency torch plasmatron 3 to generate air plasma jets with temperatures up to 4000 K. Air exhauster 9 provides constant air flow through the HFT-plasmatron 3 and impeller 7 in reactor.

During the plasma utilization process of modelling WOC dusty and steam mixture creates into the reactor. It includes metal oxides going through the centrifugal-bubbling device in wet cleaning unit for exhausted gases. At the outlet this mixture forms water suspensions from dispersed solid products included magnetic iron oxide. It's worth noting about interest of application of magnetic precipitation for dispersed solid products (with magnetic iron oxide) extraction.

Figure 4 shows experimental scheme for the comparative gravity and magnetic precipitation of dispersed solid plasma utilization products from water suspension.

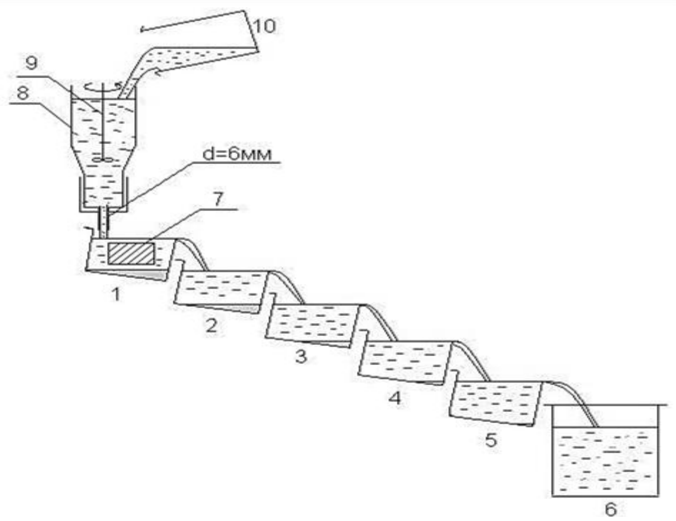


Figure 4. Experimental scheme for the comparative gravity and magnetic precipitation of dispersed solid plasma utilization products

For the precipitation of powders from water suspensions magnet M_1 (50x30x10 mm) was used. It is made from Fe-Nd-B alloy with magnetic induction 1,2 T.

Figure 5 shows the result of research gravity and joint gravity and magnetic precipitation dispersed solid plasma utilization products (with magnetic iron oxide) from water suspension.

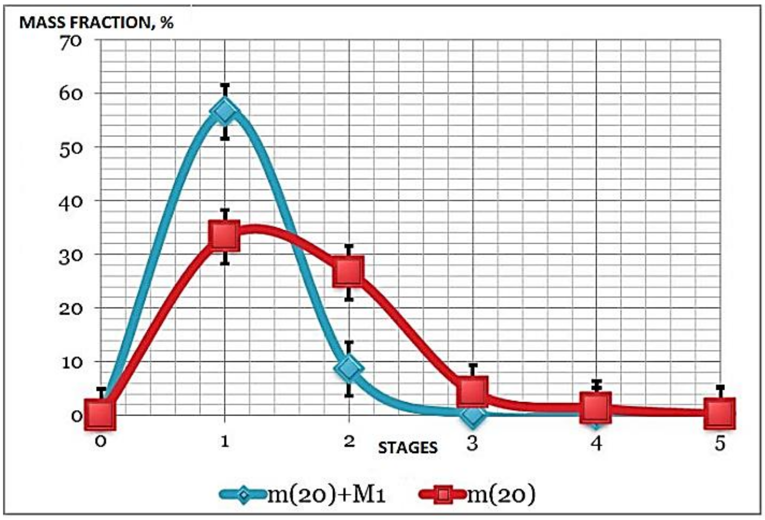


Figure 5. Comparative efficiency of gravity and joint gravity and magnetic precipitation dispersed solid plasma utilization products (with magnetic iron oxide) from water suspension

Analyzed the graph should be that when there are equal initial contents of ferriferous powders in water suspension (20 g/l) then application of magnetic precipitation considerably increases of extraction of powders from water suspensions (with magnetic iron oxide) in comparison with the only gravity precipitation. This indicates that there is possible to apply magnetic separators for effective extraction of solid SNF plasma utilization products from water suspension.

5. Conclusion

All the obtained results could be used in creating the commercial plants for complex plasma utilization of SNF processing wastes and other liquid radioactive wastes.

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