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Abstract. Graphite elements of nuclear power plants accumulate in time a small amount of uranium, radionuclide conversion elements, as well as fission products, which creates additional difficulties in the utilization of graphite masonry. We proposed a new method of processing, based on high-temperature separation in a nitrogen atmosphere. The processing consists in heating the radioactive graphite in an inert nitrogen atmosphere to a temperature of 2873 K, which creates the necessary conditions for the separation of radionuclides from carbon.

1. Introduction

Reactor graphite is applied as structural material of the moderator and the reflector in uranium - graphite reactors of channel type. Besides reactor graphite is used as the reflector blocks and fuel elements covers (fuel elements) of the high-temperature gas-cooled nuclear reactors [1].

Distinctive features of the irradiated reactor graphite as the solid radioactive waste formed after decommissioning which have wide range of radionuclides-pollutants and carbon isotope ^{14}C. The total amount of the irradiated graphite is about 230-250 thousand tons in the world, the existing ways of its processing have a number of basic shortcomings [2]. Taking into account period from 2018 to 2023 the removal from operation the urano-graphite power units of the first generation program is implemented, and since 2025 gradual power units of the second generation will be removed from the NPP [2], radioactive graphite processing problem becomes acute.

In the patent [3] the new way of reactor graphite processing in the low-temperature gas-discharge plasma conditions in the nitrogen atmosphere is suggested. It was offered on the basis of the carried-out thermodynamic modeling system of radioactive graphite – nitrogen [4, 5] with application of the TERRA complex program based on which phase distribution data and the formed compounds of radionuclides in the considered temperature intervals were obtained. The forms of existence of radionuclides in graphite and balanced system used in modeling are presented in table 1.

2. Experiments

As an example in figures 1, 2 and 3 the distributions of carbon, uranium and plutonium in a system reactor graphite-nitrogen received by means of the carried-out thermodynamic modeling are represented.

Obtained results showed (figure 1) that when heating the system to temperature of 2873K carbon completely is in the condensed phase. Further temperature increase in a system from 2873 to 3573 K causes to transition from the condensed phase to gaseous with formation of CN, C_3, C_2, C and C_2N volatile compounds. In 3573 – 4273K C_3 concentration reduction and CN, S, C_2 increase is observed.

Table 1. Forms of existence of radionuclides in graphite and balanced system.

Radionuclide in graphite	Connection type in balanced system*
^{10}Be	$BeO_{(cond)}$, B_3O_3, BeO, $Be_3N_{2(cond)}$, $BeCl_2$, BeC_2, Be, Be_4O_4, Be_2O_2
^{14}C	$C_{(cond)}$, C, CO, CO_2, CN, C_2N_2, C_2N, C_3
^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu	$PuCl_{3(cond)}$, $PuC_{2(cond)}$, $PuC_{(cond)}$, Pu, Pu^+, $PuO_{2(cond)}$, PuO_2, $PuO_{(cond)}$
^{238}U, ^{236}U, ^{235}U	$UO_2(NO_3)_{2(cond)}$, $UO_{3(cond)}$, $UO_{2(cond)}$, $U_3O_{8(cond)}$, U, $U_{(cond)}$, UCl, UCl_2, $UCl_{3(cond)}$, UCl_3, $UCl_{4(cond)}$, UCl_4, $UCl_{5(cond)}$, UCl_5, UCl_6, UO, $UO_{(cond)}$, UO_2, UO_3, $UOCl_{(cond)}$, $UOCl_{2(cond)}$, $UO_2Cl_{(cond)}$, $UO_2Cl_{2(cond)}$, UO_2Cl_2, $UO_2CO_{3(cond)}$, $NaUO_{3(cond)}$, $NaUO_{4(cond)}$, $Na_2UO_{4(cond)}$, $UC_{(cond)}$, $U_2C_{3(cond)}$, $UC_{2(cond)}$, U^+
^{36}Cl	Connections with Na, K, Cs, U, Ni, Pu, Li, Eu
^{41}Ca	Ca, $Ca_{(cond)}$, Ca_2, CaCl, $CaCl_{2(cond)}$, $CaCl_2$, $CaCO_{3(cond)}$, CaO, $CaO_{(cond)}$, $CaO_{2(cond)}$, Ca^+
^{59}Ni	Ni, $Ni_{(cond)}$, Ni_2, NiCl, $NiCl_2$, $NiCl_{2(cond)}$, $NiCl_3$, $NiCO_{3(cond)}$, NiO, $NiO_{(cond)}$, $Ni(CO)_{4(cond)}$, $Ni_3C_{(cond)}$, Ni^+
^{90}Sr	Sr, $Sr_{(r)}$, $Sr_{2(r)}$, $SrCl_{(r)}$, $SrCl_2$, $SrCl_{2(r)}$, $SrCO_3$, SrO, $SrO_{(r)}$, SrO_2
^{137}Cs, ^{134}Cs	$Cs_{(cond)}$, Cs, CsCl, $CsCl_{(cond)}$, Cs_2Cl_2, $CsClO_{3(cond)}$, $CsClO_{4(cond)}$, $Cs_2CO_{3(cond)}$, Cs_2CO_3, $CsNO_2$, $CsNO_3$, CsO, CsO_2, $Cs_2O_{(cond)}$, $Cs_2O_{(cond)}$, $Cs_2O_{2(cond)}$, Cs_2O_2, $Cs_2O_{3(cond)}$, Cs^+
^{241}Am, ^{243}Am	Am, $Am_{(cond)}$, $AmCl_{3(cond)}$, $AmO_{2(cond)}$, $Am_2O_{3(cond)}$, $AmOCl_{(cond)}$
^{150}Eu, ^{151}Eu, ^{152}Eu, ^{153}Eu, ^{154}Eu, ^{155}Eu	$EuCl_{2(cond)}$, Eu, Eu^+, $Eu_2O_{3(cond)}$, $EuO_{(cond)}$

*Notice: index (cond) - condensed phase, without an index - the gaseous phase

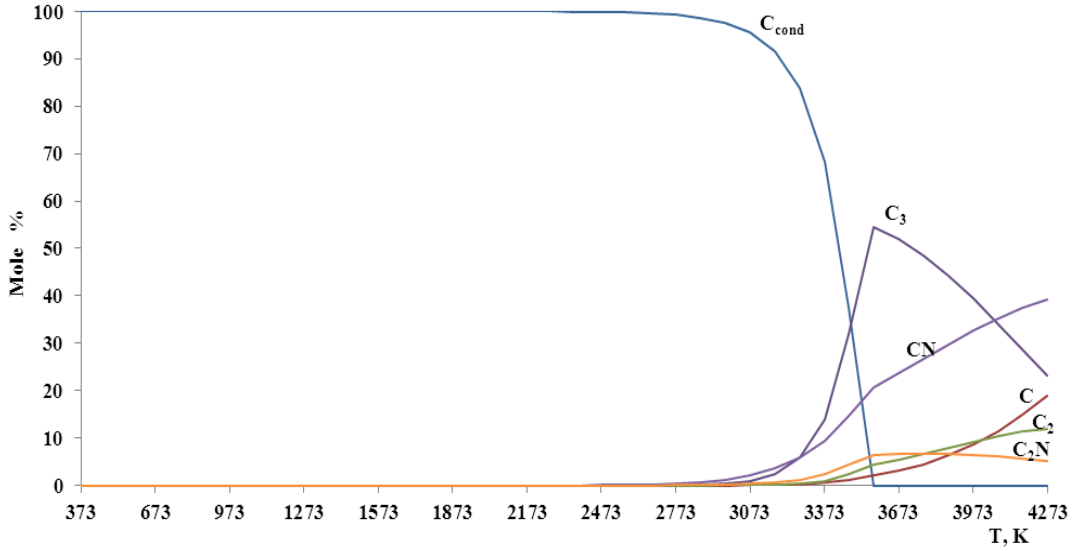

Figure 1. Uranium distribution on phases.

The uranium balance in the considered system shows (figure 2) that up to 873K uranium is in the condensed phase in the form of the UC, UC_2, U_2C_3 ,UCl_3 compounds. Further heating of a system from 873K to 2073K results in formation of gaseous chlorides of UCl_3 and UCl_4 uranium. At temperatures from 2473K to 2773K concentration of the condensed compounds decreases and also couple U and U^+ are formed. At higher temperatures 2773 K there is a transition of gaseous uranium U to uranium U^+ ion. The balances of all radionuclides accumulated in radioactive graphite were similarly analyzed.

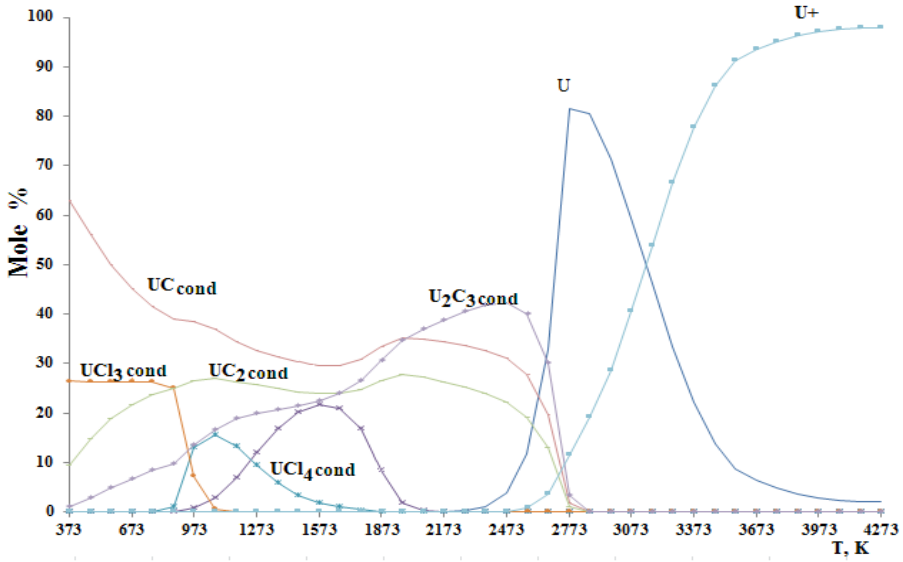

Figure 2. Uranium distribution on phases.

Plutonium distribution on equilibrium phases is presented in figure 3. In the temperature range from 373 to 1073K plutonium is in the condensed phase in the form of $PuCl_3$ compound. At temperatures 1073 - 1573 K concentration of $PuCl_3$ declines and the amount of the condensed plutonium carbides as PuC_2 and PuC increases. Further temperature rise from 1573 to 2373 K causes the plutonium transition to a gaseous phase in the form of Pu vapors. In the range of temperatures 2373 – 4273 K there is a transition of gaseous plutonium Pu to a plutonium Pu^+ ion with the formation of an e^- electronic cloud.

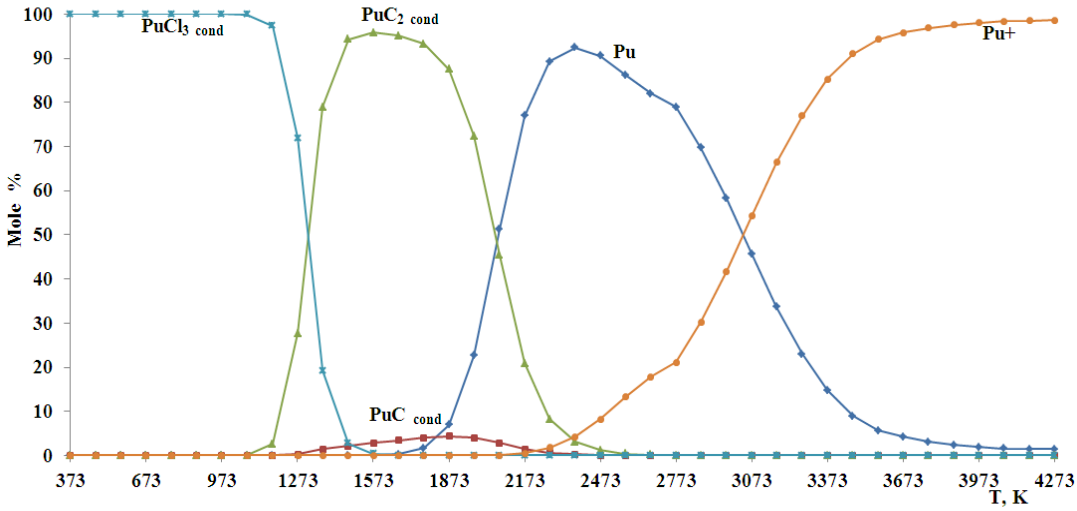

Figure 3. Plutonium distribution on phases.

Data of all radionuclides distribution balances comprising reactor graphite are similarly obtained and analyzed, these results are represented in the general table 2**.**

Table 2. Distribution of elements in the temperature ranges in the conditions of reactor graphite heating.

Element	Temperature range of phase distribution (Formed compounds)		
	Only condensed phase	Intermediate interval (two phases in system)	Only gaseous phase (vapours)
Carbon (^{12}C, ^{14}C)	2873 < (C)	2873 - 3573	> 3573 (C_3, CN, C, C_2)
Uranium	2373 < (UC, UCl_3, UCl_4, U_2C_3, UC_2, UC_2)	2373 - 2873	> 2873 (U, U^+)
Plutonium	1673 < ($PuCl_3$, PuC_2, PuC)	1673 - 2573	> 2573 (Pu, Pu^+)
Americium	1373 < (Am)	1373 - 2573	> 2573 (Am)
Europium	1473 < ($EuCl_2$)	1473 - 1873	> 1873 (Eu, Eu^+)
Strontium	973 < ($SrCl_2$)	973 - 1373	> 1373 ($SrCl_2$, Sr, Sr^+, SrCl)
Cesium	673 < (CsCl)	673 - 973	> 973 (CsCl, Cs^+)
Nickel	1273 < (Ni, Ni_3C)	1273 - 1473	> 1473 (Ni, NiCl, Ni^+)
Chlorine	773 < (UCl_3, $CaCl_2$)	773 - 1273	> 1273 (UCl_4, UCl_3, Cl, $CaCl_2$, $BeCl_2$)
Beryllium	873 < (Be_3N_2)	873 - 1073	> 1073 ($BeCl_2$, BeC_2, Be)
Calcium	973 < ($CaCl_2$)	973 - 1373	> 1373 ($CaCl_2$, CaC, Ca, Ca^+)

3. Conclusion

Thus reactor graphite processing in low-temperature gas-discharge plasma results in the following when reactor graphite powder with sizes of fractions no more than 1-2 mm is loaded into the electric arc furnace that allows intensifying the proceeding processes considerably. In the inert nitrogen atmosphere under the influence of the electric arc furnace (low-temperature gas-discharge plasma) graphite heats up to working temperatures of 2873 – 2923 K creating necessary conditions of radionuclides transition to gaseous phase except carbon and its isotope ^{14}C along with it the vacuum system removes the formed volatile compounds from the working chamber of the furnace and passes them via the filters system for their catching.

4. References

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