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Probe diagnostics of $^3He + UF_6$ plasma, generated in the core of nuclear reactor WWW–K

Abstract. In the present paper experimental results of probe measurements in nuclear induced plasma in the presence of negative ions are presented and used to define the ratio of electrons and negative ions directly taken from electrostatic probe volt–ampere characteristic (VAC) inserted in the $^3He+UF_6$ mixture.

The nuclear induced plasma in the core of nuclear reactor was created by the following nuclear reaction $^3He + n \rightarrow p + T + 0.76 MeV$. The quantitative analysis is also developed to get detailed explanations of the probe diagnostics technique and to define the density of negative ions in gas mixtures like $^3He + UF_6$ where the electrons and negative ions are presented in unknown proportion. Experimental study of uranium hexafluoride plasma of high pressure is an important part of a comprehensive problem to get efficient direct conversion of nuclear energy of fission fragments not only into the heat transfer but also into some other different forms of energy. Probe diagnostics of uranium hexafluoride in the WWR–K is a unique experiment and has been done for the first time in the former Soviet Union [1].
Key words: helium, uranium hexafluoride, mixture, probe diagnostics, nuclear energy, fission fragment.

Introduction

Commonly known that electrostatic probe is used as a primary diagnostic tool in the measurement of the local parameters of the ionized gas in a variety of medium [2–3], such as the electrical discharge and afterglow, the ionization boundary region behind the shock waves, flames, MHD generators, plasma jet, as well as atmospheric and space plasmas. Despite its limited area of application the probe techniques of experimental measurements are very successful and rapidly developed in recent years. In some special cases like plasma diagnostics in the core of active of nuclear reactor the probe diagnostic methods is left as the only one possible experimental technique which makes possible to extract nuclear induced plasma property information (figure 1). Electrostatic probe represents substantially a metal electrode placed in the diagnostic cell, inserted in the active zone of nuclear reactor and within which the tested mixture is uploaded .The probe diagnostics experimental set in nuclear reactor is schematically presented on the Figure 2 [4, 8].
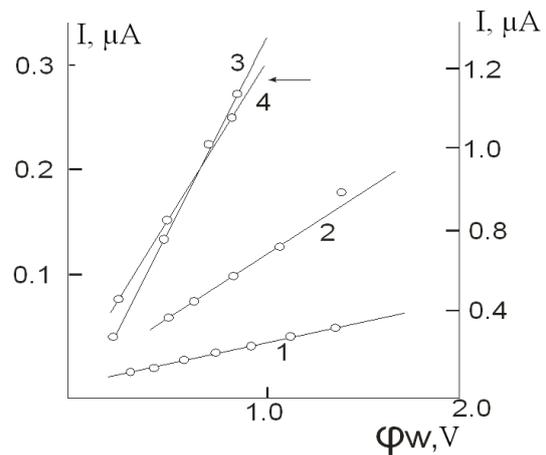
1–Helium –3 plasma +UF6 admixture, 1–Helium–3 plasma.

Figure 1 – Volt ampere characteristics of electrostatic probes in the core of nuclear reactor.

The plasma apparatus is relatively simple, however, the theory of electrical probes complicated by the fact that the probes are boundary surface with respect to the plasma, and the equations describing

the behavior of the plasma near the interface are nonlinear [2]. The plasma created in the active zone of a nuclear reactor has a number of specific features. These include, in particular, inseparably related problems: chemical aggressiveness and toxicity of raw materials, the impossibility of direct contact with the experimental set due to induced radioactivity, making all of the experimental apparatus is only a one–time use as well as the obligatory remote control of the experiment set due to the irreversible structural changes in the measurement and diagnostic devices connected with strong radiation.[4] Plasma, created by fission fragments was initially described in the following papers by Leffert C. B, Reese D. B., Nguyen D. H., Grossman L.M. and Guyot J. C., Miley G. H., Verdeyen J. T. [5–7].
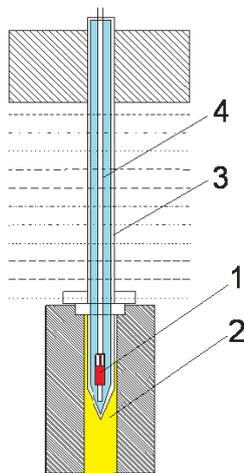
1 – Vial of test mixture; 2 – Reactor core; 3 – Diagnostic channel; 4 – Signal wires

Figure 2 – Experimental setup for probe diagnostics of the plasma gas mixtures in the radiation field of a stationary nuclear reactor.

Experimental volt ampere characteristics analysis

The test mixture of $^3He + UF_6$ of the gas sample inserted in the flux of thermal neutrons absorbs thermal neutrons creating highly energetic particles, which cause in its own turn the ionization of the working medium through the following channel:

$$^3He + n \rightarrow \; ^3H + p + 0.76 Mev, \quad (1)$$
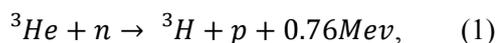
where He – helium, n – neutron, p – proton, H – hydrogen.

Reaction cross section (1) for thermal neutrons is 5400 barns and increases as its energy varies pro-

portional to $E^{-0.5}$. The energy released distributed among the proton and triton as follows: the nucleus of tritium $-578\,kev$ and $198\,kev$ proton energy. The input power of the reaction ³He(n,p)T per unit volume, estimated by the following expression:

$$P = 1.65 * \Phi * [\,^3He] \frac{W}{sm^3}, \quad (2)$$

where Φ – neutron flux.

The experimental set of probe diagnostics of the plasma formed by the products of nuclear reactions, schematically shown in figure 2.

The diagnostic channel into which the test cell inserted is made of steel pipe. The diagnostic cell has the shape of a cylinder with a diameter of 40 mm. At the bottom of the cell special tip was made to fill the gas mixture that was to be studied. In the center of the upper base of the cell the probes of different geometry configurations were welded, sealed and mounted to the signal wires. The probes are mounted on a ceramic holder. Non–operating part of the probe was protected from contact with plasma by insulator made of the quartz tubes. Pumping, heating and filling cells with gas mixture carried out with a high–vacuum unit [8].

In the present paper, the detailed quantitative consideration of this problem is made and compared with experimental data realized by one of the authors [7].

At any surrounding probe surface we have:

$$\oint \overrightarrow{J^+} d\vec{s} = I_p^+, \quad (3)$$

where I_p^+ – current of positive ions on the probe.

$$\oint \overrightarrow{J^e} d\vec{s} = I_p^e, \quad (4)$$

where I_p^e – current of electron on the probe.

We also accept that at any point:

$$I^e + I^+ = I_p. \quad (5)$$

From experimental curve, we may notice that ion and electron branches of VAC at some definite region are linear and the slope might be exactly taken out from the experimental VAC.

In the charged volume layer, we may state that:

$$div J^+ = S, \quad (6)$$

and the radius of the layer is equal to:

$$r_0^+ = r_p(1 + \frac{I_p^+}{4\pi e S r_p^3})^{\frac{1}{3}}, \qquad (7)$$

where e – electron's charge.

From this we may obtain, that:

$$I_p^+ = 4\pi e r_0^+ n^+ b^+ \varphi_p^{negative}, \qquad (8)$$

where $\varphi_p^{negative}$ – probe's potential, b^+ – mobility coefficient, r_0^+ – length charge layer.

To get concentration of positive ions in plasma we come to the following:

$$n^+ = \frac{\tan \alpha^+}{4\pi e b^+ r_0^+}, \qquad (9)$$

where

$$\tan \alpha^+ = \frac{\Delta I^+}{\Delta \varphi^{negative}}. \qquad (10)$$

The presence of negative ions strongly affects on the slope of electronic part of the probe VAC, in some equals to the same value of the ion branch, and become symmetric.

If we shall denote the ratio of electrons concentration to concentration of positive ions in plasma as:

$$\delta = \frac{n^e}{n^+}, \qquad (11)$$

and proceed our evaluations for positive values of the probe potential then for electronic part of the probe (Figure 3) VAC we get:

$$I_p^e = 4\pi e r_0^e n^e \varphi_p^{positive}. \qquad (12)$$

Generally we write:

$$\delta = \frac{b^+}{b^e} \frac{\tan \alpha^e}{\tan \alpha^+} \frac{r_0^+}{r_0^e} \qquad (13)$$
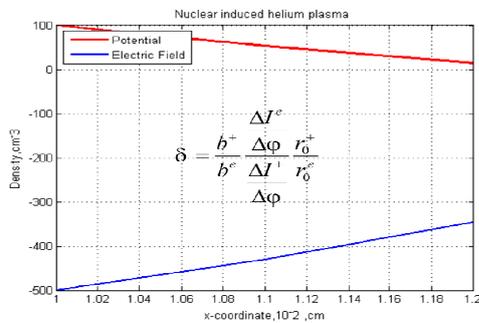
Figure 3 – Numerical calculations of potential and electric field in the spherical probe charged layer.

This parameter may change from $1 - 10^{-2}$ and depends on the concentration of any admixture like UF^6 leading to the formation of negative ions. On the figure 4 it is clearly shown the electron's and ion dependence from neutron flux. Electron's density is linear function from neutron flux intensity as well as ion's density is proportional to its square root.
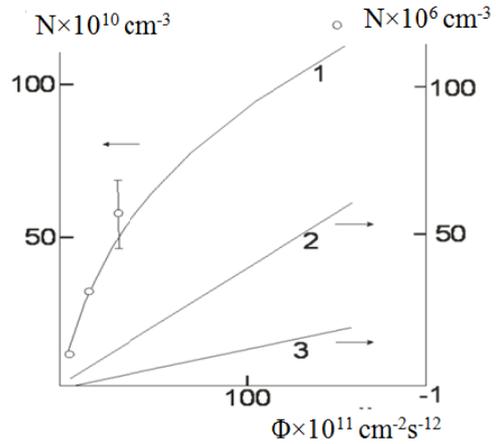
– calculated; o – experiment; 1 – UF_6^+; 2 – n_e; 3 – UF_5.
Figure 4 – Densities of charged particles dependence from the thermal neutron flux density.

The figure 5 shows the linear dependence of ion current from probes potential and confirms that the voltage drop is lying in the charged layer. From (6) and Poisson equation for charged layer the potential obeys to the following differential equation:

$$\varepsilon \omega^+ \theta[\frac{\partial^3 \Psi}{\partial \xi^3} \frac{\partial \Psi}{\partial \xi} + [\frac{\partial \Psi}{\partial \xi}]^2] = 1, \qquad (14)$$

where

$$\xi = \frac{r}{r_p}, \theta = \frac{T^e}{T^+}, \omega^+ = \frac{D^+ n_\infty^+}{S}, \varepsilon = (\frac{r_d}{r_p})^2, \Psi = \frac{\phi}{\frac{kT^e}{e}}.$$

Boundary conditions are as follows:

$$\Psi(1) = \Psi_p, \Psi''(1) = 0. \qquad (15)$$

Electric field in the charged layer

$$\frac{\partial \phi}{\partial r} = \sqrt{\beta} r_0 (1 - \xi^2{}_0)^{\frac{1}{2}}, \beta = \frac{4\pi e S}{b^+}, \qquad (16)$$

where S – ionization rate.

$$r_0 = r_p + \left(\frac{36}{25}\frac{\varphi_p^2}{\beta}\right)^{\frac{1}{4}}. \qquad (17)$$

For the interpretation of the VAC it is necessary to evaluate the ion composition of the plasma. In the model the following elementary processes are taken account. High–energy fission fragments, which are formed by the interaction of thermal neutrons with ^{235}U, characterized by an initial energies lying in the range 50–115 MeV, the initial charge of +16e to +24e and weights from 70 to 160 amu. Under the influence of high uranium fission fragments in the gas a cascade of fast electrons is formed. These electrons and fission fragments produce ionization of molecules

$$UF_6 + (ff) \rightarrow UF_6^+ + e + (ff)', \qquad (18)$$

$$UF_6 + (ff) \rightarrow UF_6^* + (ff)', \qquad (19)$$

$$UF_6 + (ff) \rightarrow UF_6 + F + e + (ff)', \quad (20)$$

$$UF_6 \rightarrow UF_5 + F, UF_6^* \rightarrow UF_6 \rightarrow UF_4 + F_2, \quad (21)$$
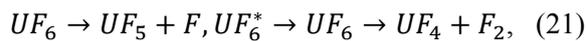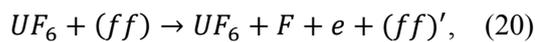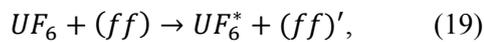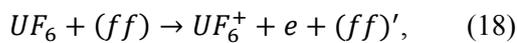
where ff – fission fragments, UF – uranium hexafluoride, F – fluorine.

The rates of ionization and excitation per unit volume, estimated by the formula:

$$J^- = -D_a^- \frac{d}{dx} N^-, \qquad (22)$$

where D_a^- – unbipolar diffusion coefficient of negative ions.

$$S_1 = \Phi q_{fis} EN/U_i, \qquad (23)$$

where Φ – thermal neutron flux, q_{fis} – nuclear reaction cross section, E – energy division, N – concentration of fissile material, U_i – the energy cost of a corresponding product in this reaction.

The total cross section of the positive ions in the plasma of uranium hexafluoride in collisions of electrons with gas molecules, since the threshold to 1 keV. In the same paper we present plots of the total current of negative ions, which are produced by collisions of low–energy electrons (E ≤ 10 eV) with a molecule of UF6, and is the dominant ion UF_5 −. Total current peaks are observed in the energy range (2.1±0.1), ~ 7 and ~ 11 eV. Formation UF_5 − is realized in accordance with the reaction:

$$e + UF_6 \rightarrow UF_5^- + F. \qquad (24)$$
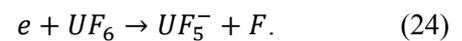
Same processes of electron attachment are leading to the formation UF_6− characterized by a rather low rate. It is considered that leads to the emergence of UF_6 – exchange reaction:

$$UF_5^- + UF_6 \rightarrow UF_6^- + UF_6. \qquad (25)$$
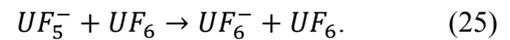
It is known that under the influence of radiation complex molecules substances are destroyed. The most significant destructive effect produces fission fragments. UF_6 molecules under the influence of the radiation of the reactor will be destroyed at the lower fluorides and fluoride. Under the assumption that the dissociation of UF6 molecules is via

$$UF_6 \rightarrow UF_5 + 1/2F. \qquad (26)$$
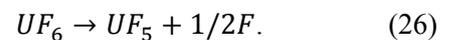
Equation 26 determines the rate of destruction of UF_6− per unit of power. Within the measurement error rate of radiolysis of UF$_6$ molecules is independent of pressure and dose rate and amounts to 0.28 mol / kWh of energy absorbed or (0.8±0.1) molecules / 100 eV. However, it is known that uranium fluorides are relatively easy might be fluorinated with increase of gas temperature:

$$UF_5 + F + M \rightarrow UF_6 + M, \qquad (27)$$

$$UF_4 + F_2 + M \rightarrow UF_6 + M, \qquad (28)$$
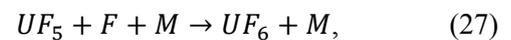
where M – the third particle.

According to [9], uranium hexafluoride is thermally stable compound. Decomposition of UF_6 into fragments can be observed at temperatures above 2200 K.

The above mentioned positive and negative ions, electrons, and the dissociation products interact strongly with one another.

The constants of plasma–chemical reactions for UF_6 might be estimated and taken from [8 and 9].

$$F + F + M \rightarrow F_2 + M. \qquad (29)$$

$$e + F_2 \rightarrow F^- + F. \qquad (30)$$

$$e + UF_6^+ \rightarrow \frac{UF_5 + F}{UF_4 + F_2}. \qquad (31)$$

$$e + UF_6^+ + M \rightarrow UF_6 + M. \qquad (32)$$

$$F^- + UF_6 \rightarrow UF_6^- + F. \qquad (33)$$

$$UF_6^+ + UF_6^- \rightarrow 2UF_6. \qquad (34)$$

$$UF_6^+ + UF_6^- + M \rightarrow +UF_6 + UF_5 + F + M. \qquad (35)$$

$$UF_6^+ + F^- + M \rightarrow UF_6 + F + M. \qquad (36)$$

$$UF_6^+ + F^- \rightarrow UF_6 + F. \qquad (37)$$
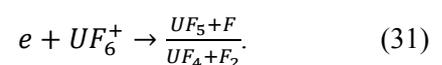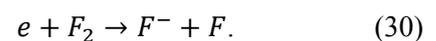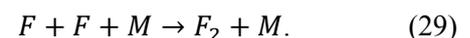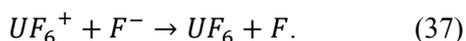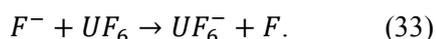
The system of equations described mathematical model was solved numerically. To match the experimental conditions to study plasma probe method in the submitted calculations pressure UF_6 assumed to be 20 Torr, the thermal neutron flux varies from 3 to $1.5 * 10^{11} * 10^{13}$ neutrons cm^{-2}s^{-1}. Further, in view of the fact that the probe measurements were carried out for 2–3 hours, we believe that under these conditions the concentration of uranium hexafluoride, UF_6 in (23) is equal to the original content. As an example, in Table 1 shows the results of calculations for the conditions of the experiments.

As it is shown in Table 1, main negative ions in the mixture are UF_6^- ions . Positive ions concentration is the concentration of UF_6^+ ions (up to five digits). The density of UF_6^- determined by the ionization and recombination reactions .The electron's density is negligibly small. The calculation results show that the ratio of the electron density to the total concentration of negative ions is of the order of 10^{-4}. The electron's density is also determined by the ionization and attachment to molecules of UF_6. The numerical calculations confirm that the concentration of negative ions is much greater than the concentration of electrons and corresponds to the results obtained from the analysis of the CVC. Furthermore the concentration of negative ions proportional to the square root of the value thermal neutron flux, i.e. $\sim \sqrt{\Phi}$, the electron density is linear proportional to the neutron flux, i.e. $\sim \Phi$.

Table 1 – Density of uranium hexafluoride plasma ions

n_e	UF_6^+	UF_6^-	UF_5^-	F^-
$5.9*10^6$	$2.6*10^{11}$	$2.6*10^{11}$	$2.1*10^6$	$4.2*10^4$
F	F_2	UF_5	UF_4	
$7.7*10^{13}$	$7.7*10^{16}$	$3.9*10^{14}$	$4.0*10^4$	

Conclusions

The experimental VAC in some regions is linear function of probe's current and its potential and the slopes of the curve might be used to measure the electrons and negative ions ratio. On figure 5 the measured current–voltage characteristics of the electric probe in the plasma of uranium hexafluoride are presented. It was established that the CVC corresponding reactor power level of 100–200 kW are symmetric. From the symmetry of the characteristics we may come to the following conclusions: firstly, the contribution of the electrons in the plasma conductivity is negligibly small, i.e., $n^-\mu^- = n_e\mu_e$, however, since $\mu^- \ll \mu^e$, then it comes to be that $n^- \gg n^e$.
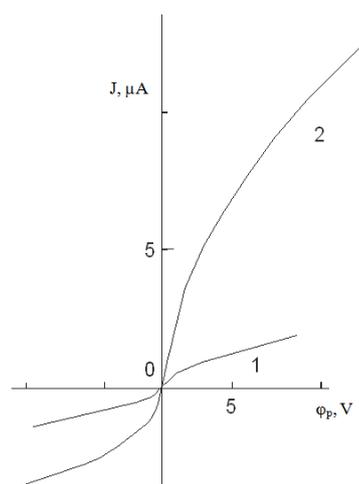
Figure 5 – Voltage characteristics of uranium hexafluoride at different power levels in the sample ^{3}He+UF_6 , irradiated by neutron flux of nuclear reactor WWR–K $(1–1.10^{12}$ cm^{-2}s^{-1}, 2–110 ^{13}cm^{-2}s^{-1}).

The kinetic model of plasma UF6 and numerical calculation of ionic composition of the plasma also confirms this relationship. From the analysis of the results of calculations we may state that the ion concentration $UF_6^- = UF_6^+$ (up to the fifth sign) and $UF_6^+(UF_6^-)\sim\sqrt{\Phi}$, $n_e\sim\Phi$, (Φ – thermal neutron flux). The experimental and calculated values of the concentrations of positive ions are in a satisfactory agreement.

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