

Research Article

Emission of Noble Gases Binary Mixtures under Excitation by the Products of the ${}^6\text{Li} (n, \alpha) {}^3\text{H}$ Nuclear Reaction

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The luminescence of Kr-Xe, Ar-Kr, and Ar-Xe mixtures was studied in the spectral range 300–970 nm when excited by ${}^6\text{Li} (n, \alpha) {}^3\text{H}$ nuclear reaction products in the core of a nuclear reactor. Lithium was deposited on walls of experimental cell in the form of a capillary-porous structure, which made it possible to measure up to a temperature of 730 K. The temperature dependence of the radiation intensity of noble gas atoms, alkali metals, and heteronuclear ionic noble gas molecules was studied. Also, as in the case of single-component gases, the appearance of lithium lines and impurities of sodium and potassium is associated with vaporization during the release of nuclear reaction products from the lithium layer. The excitation of lithium atoms occurs mainly as a result of the Penning process of lithium atoms on noble gas atoms in the 1s states and subsequent ion-molecular reactions. Simultaneous radiation at transitions of atoms of noble gases and lithium, heteronuclear ion molecules of noble gases allows us to increase the efficiency of direct conversion of nuclear energy into light.

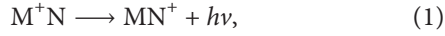
1. Introduction

The study of optical (laser and spontaneous) radiation from a nuclear-excited plasma is interesting for developing a method of energy output from a nuclear reactor, systems for monitoring the parameters of nuclear reactors [1–3], as well as for the creation of one of diagnostics of high-temperature plasma in fusion reactors [4]. The direct excitation of gaseous media is carried out, as a rule, by the products of nuclear reactions with thermal neutrons of a nuclear reactor: ${}^3\text{He} (n, p) {}^3\text{H}$, ${}^{10}\text{B} (n, \alpha) {}^7\text{Li}$, ${}^{235}\text{U} (n, f) \text{F}$, or others [3]. The gaseous medium must contain ${}^{235}\text{U}$, ${}^3\text{He}$, or ${}^{10}\text{B}$, or a compound with these isotopes is applied to the walls of chamber. In [5, 6], to study the luminescence of gases in the core of a nuclear reactor at temperatures of 310–730 K, the products of ${}^6\text{Li} (n, \alpha) {}^3\text{H}$ nuclear reaction were used. The

relatively large mean free path of tritium nuclei in lithium and gaseous media makes it possible to excite large volumes of gases and provide a larger amount of power deposited in the gas in comparison with the reaction products with ${}^{10}\text{B}$. An increase in temperature of gaseous medium to ~500 K led to the appearance in the emission spectra of lines of alkali metals, lithium, and impurities of sodium and potassium. The appearance of these lines is associated with the evaporation of lithium under emission of α -particles and tritium nuclei from the deposited layer [6]. In single-component noble gases, the excitation of lithium atoms occurs as a result of the Penning process on metastable noble gas atoms (atoms in 1s states) [6]. In the visible region, intense radiation from lithium atoms is added to the radiation at the $2p$ -1s transitions of noble gas atoms (Paschen's notation). In this case, the appearance of radiation at lithium transitions

does not affect the intensity of $2p$ - $1s$ transitions of noble gases, since the excitation is transmitted to lithium atoms from the lower $1s$ levels. As a result, the efficiency of converting nuclear energy into light increases considerably.

Molecular bands observed in the emission spectra of paired mixtures of noble gases were identified [7] as transitions between the states of heteronuclear ion molecules:

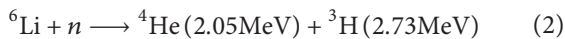


where the molecular states of M^+N asymptotically correspond to the states of $M^+ + N$ and MN^+ to the states of $M + N^+$; here M , N are noble gas atoms, and N is a heavier atom. A total of nine transitions between the states M^+N and MN^+ are possible; five of them were observed in the electric discharge. Transitions from the general level dissociating to $M^+ (^2P_{1/2})$ и $N (^1S_0)$ are denoted by A and D , and transitions from the $M^+ (^2P_{3/2})$ N states are denoted by B , C , and E [7]. In an electric discharge, the intensity of the five bands is comparable in magnitude. When excited by ionizing radiation, bands A and D dominate sharply [8–11]; these bands have a high radiation efficiency. In [12], when the mixture $Kr : Xe = 1 : 1$ at a total pressure of 111 kPa was excited by the decay products of the ^{210}Po isotope, the radiation efficiency in the 491 nm band of the KrXe^+ ion molecule was determined to be 11% of the α -particles power deposited in the gas.

In this work, we studied the luminescence of Ar-Kr, Kr-Xe, and Ar-Xe mixtures excited by the products of $^6\text{Li} (n, \alpha)^3\text{H}$ nuclear reaction. The temperature dependence of radiation at transitions of noble gas atoms, alkali metal atoms, and heteronuclear ionic noble gas molecules has been studied. The results are of interest from the point of view of increasing the efficiency of conversion of nuclear energy into light.

2. Experimental Setup

The studies were carried out at the IVG.1 M stationary research nuclear reactor [13, 14] at a thermal neutron flux density of up to $1.4 \cdot 10^{14}$ n/cm²s. Ionization and excitation of the gaseous medium were carried out by the products of a nuclear reaction:



Promising for use as a surface source of charged particles is lithium in the liquid phase. Therefore, in comparison with lithium in the solid phase, surface of the source due to convective mixing of liquid lithium will constantly update itself, and the products of lithium interaction with the gas phase (oxides, hydrides, carbides, nitrides, etc.) from the surface will be removed deep into the source. In [5], lithium was applied to the walls of the experimental cell by heating up to 570 K, and liquid lithium in a vacuum spread evenly over the surface and was retained due to the surface tension force. In present work, the stabilization of liquid lithium in the source is achieved through the use of a capillary-porous structure (CPS) as a stabilizing matrix [15]. The CPS properties are based on the dependence of the capillary

pressure p_c in the channels on the radius R of the liquid metal meniscus on the CPS surface: $p_c \sim (1/R)$. Thus, capillary forces provide stabilization and uniform distribution of liquid metal.

The scheme of the experimental device with lithium CPS is shown in Figure 1. The experimental device is made of stainless-steel pipe with a length of 900 and an inner diameter of 20 mm. At the top of the device, it is plugged with a flange with an optical vacuum inlet (1).

To ensure the necessary temperature conditions, the experimental device was equipped with two ohmic heaters (2, 3) mounted on the housing and on lithium part of the cell. The temperature of walls of the experimental device was controlled using four chromel-alumel thermoelectric converters (4) located at 4 points along the height of the outer surface.

To create a lithium layer, a woven mesh made of stainless steel was placed inside the experimental cell. The dimensions of the porous cell were 0.3×0.3 mm and the thickness was 0.1 mm. Lithium of 1.55 g was loaded into the chamber, and in horizontal position of the experimental device, heating was carried out to 570 K in vacuum, followed by heating in purified argon atmosphere to 730 K for 30 min and to 1030 K for 15 min. After all the above procedures, the cell heating was turned off, and it cooled naturally. When the temperature reached 520 K, argon was pumped out from the experimental device and, under continuous pumping conditions, the cell cooled to room temperature. Natural lithium (7.5% ^6Li) with an impurity content of less than 0.1% was used. The height of the deposited lithium layer (5) was 200 mm.

The experimental device was loaded into the dry experimental channel of reactor; height of the core was 800 mm. At different startups of the reactor, the middle of the lithium layer was located in the center of the reactor core either 100 or 150 mm above the center, which corresponded to the average thermal neutron flux density (F) $1.4 \cdot 10^{14}$, 10^{14} , and $5 \cdot 10^{13}$ n/cm²s, respectively. The output of light radiation was carried out using a collimator with a quartz lens (6), placed inside the experimental device and connected through an optical vacuum input with a fiber optic cable (7). The light from the experimental device through the 10-meter-long fiber passed to the input of the QE65Pro (Ocean Optics) spectrometer located in the reactor hall. The luminescence spectra were recorded in the region of 300–970 nm with spectral integration times of 0.01–10 s, and the resolution of the spectrometer was 1.5 nm.

The measurements were carried out at a constant thermal power of the reactor of 6 MW. The experimental device was heated by radiation heating of a steel pipe, energy release in a lithium layer, and ohmic heating. The temperature of the lithium layer was controlled by changing the power of the heater, and if necessary, a greater decrease in temperature by changing the nitrogen flow rate with external blowing. The volume of the experimental chamber was degassed in the reactor core at a temperature of 410 K and was washed several times with the test gas before filling; gases with an impurity content of less than 0.001% were

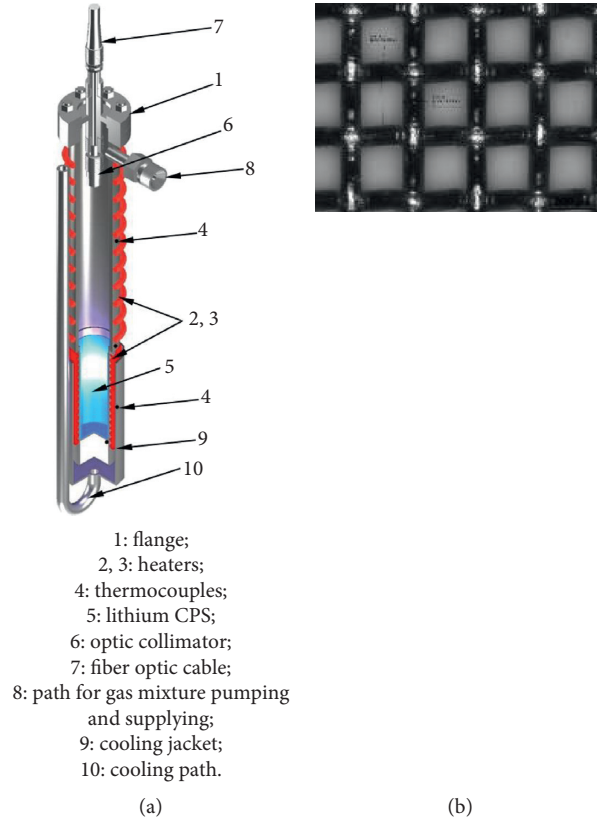


FIGURE 1: The experimental device with lithium CPS. (a) 3D view of the experimental device with (b) enlarged image of the CPS matrix lithium CPS.

used. Gas mixtures were preliminarily prepared in separate cylinders. For pumping and inlet of gas, the vacuum system of the Liana experimental bench was used [13].

3. Results and Discussion

3.1. Atomic Spectra. The temperature dependence of luminescence in Kr (51 kPa)-Xe (51 kPa), Ar (48 kPa)-Kr (48 kPa), and Ar (90 kPa)-Xe (10 kPa) mixtures was studied; the partial gas pressure is given for a temperature of 410 K. In each mixture, radiation occurs at the $2p-1s$ transitions of the heavier gas (Figure 2).

When α -particles and tritium nuclei pass through a gaseous medium, ions and secondary electrons are formed; these electrons also ionize and excite gas atoms. The main processes leading to the emission of noble gas lines in a Kr-Xe mixture (M is the third particle, krypton or xenon atom) are as follows:

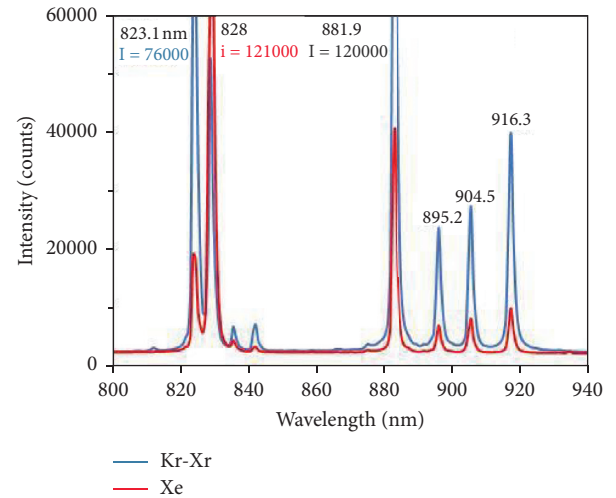
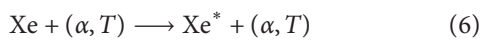
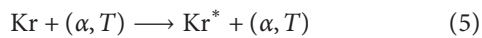
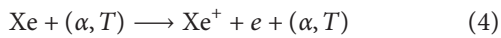
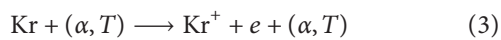
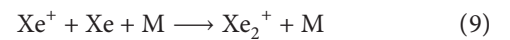
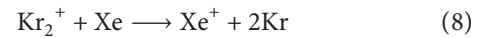
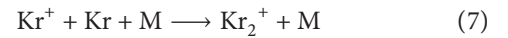
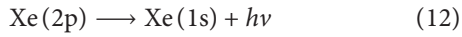
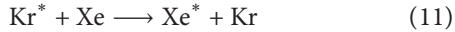
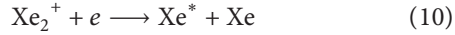


FIGURE 2: Emission spectra of xenon (36 kPa) and Kr-Xe mixture at a temperature of 413 K $F = 10^{14}$ n/cm² s. Spectrometer integration time: 300 ms (Kr-Xe), 75 ms (Xe).





As a result of dissociative recombination of the molecular ion of a noble gas, atoms in 3d and 2p states are predominantly formed [16]; 2p levels are also populated as a result of transitions from 3d levels. Possible transitions from higher levels are outside the sensitivity range of the spectrometer. The emission spectrum of the Kr-Xe mixture differs markedly from the luminescence spectrum of pure xenon (see Figure 2). This is connected not only with some difference in the processes of population of 2p levels of xenon, but also with a significant difference in the values of the quenching rate constants of 2p levels of xenon by krypton and xenon atoms [17]. This is especially noticeable when comparing the intensity of the lines of 2p5 and 2p6 xenon levels. The rate constant for quenching the 2p5 level of xenon with krypton is $13.2 \cdot 10^{-11} \text{ cm}^3 \text{ s}^{-1}$, and the quenching rate for xenon is $0.59 \cdot 10^{-11} \text{ cm}^3 \text{ s}^{-1}$. The 2p6 level, on the contrary, is more strongly quenched by xenon ($10.1 \cdot 10^{-11} \text{ cm}^3 \text{ s}^{-1}$) than krypton ($2.8 \cdot 10^{-11} \text{ cm}^3 \text{ s}^{-1}$).

As the temperature rises, lines of lithium, sodium, and potassium appear (Figure 3).

The processes of the appearance of radiation on alkali metal lines in the case of single-component noble gases are described in [6]. When α -particles and tritium nuclei are released from the lithium layer, lithium vapors are formed at a partial pressure far exceeding the saturated vapor pressure. As a result of plasma-chemical processes in a gas, lithium ions are formed, mainly as a result of the Penning process on noble gas atoms in the 1s states. Dissociative recombination of molecular lithium ions leads to the appearance of lines of lithium atoms in the spectrum. Similar processes occur with sodium and potassium vapor. The temperature dependence of the radiation intensity on atoms lines of noble gases and alkali metals is shown in Figures 4 and 5 for the Kr-Xe mixture and Figure 6 for the Ar-Kr mixture.

The intensity of krypton and xenon lines initially decreases with temperature, as well as in the case of single-component gases, and then again increases at temperatures above 500 K. The increase in intensities of the Kr and Xe lines is apparently associated with the dissociation of the molecular ions $\text{Ar}^+ \text{Xe}$ and $\text{Kr}^+ \text{Xe}$, with participation of the formed Ar^+ (Kr^+) ions in the plasma-chemical processes in the mixture. Also, as in the case of a single-component noble gas [6], the appearance of intense radiation of alkali metal atoms has practically no effect on the intensity of the lines of 2p-1s transitions of a heavier noble gas. Therefore, the main channel for transferring excitation to alkali metal atoms is the Penning process on noble gas atoms in 1s states.

The intensity of the lithium and sodium lines sharply increases at a temperature of the lithium layer above 600 K. The intensity of the resonance lines of lithium (670.4 nm, two lines are not resolved by the spectrometer) and sodium (589.5 and 590.0 nm are also not resolved) begins to

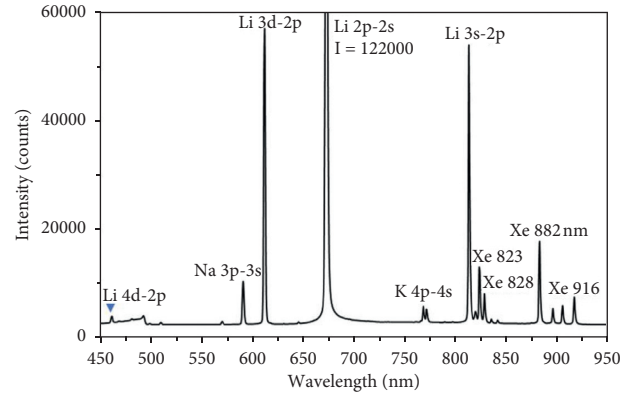


FIGURE 3: The emission spectrum of the Kr-Xe mixture at a temperature of 673 K $F = 10^{14} \text{ n/cm}^2 \text{ s}$. Integration time is 30 ms.

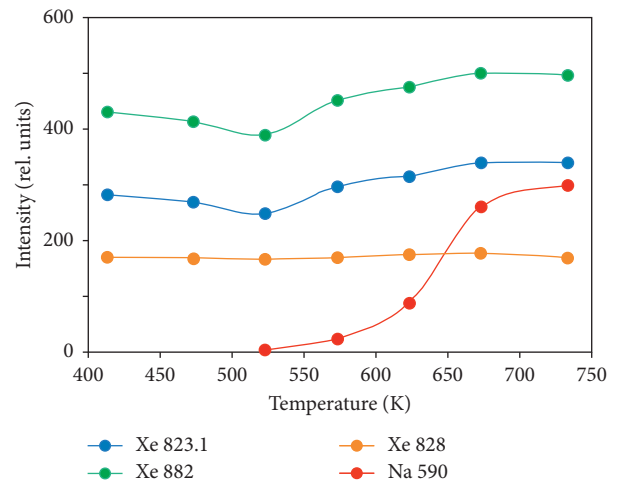


FIGURE 4: Dependence of intensity of xenon and sodium lines in the Kr-Xe mixture on the temperature of the lithium layer, $F = 10^{14} \text{ n/cm}^2 \text{ s}$.

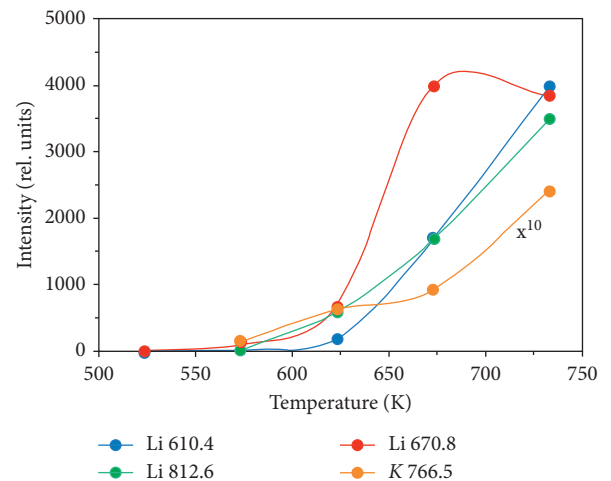


FIGURE 5: Dependence of intensity of lithium and potassium lines in the Kr-Xe mixture on the temperature of the lithium layer, $F = 10^{14} \text{ n/cm}^2 \text{ s}$. The intensity of potassium lines increased by 10 times.

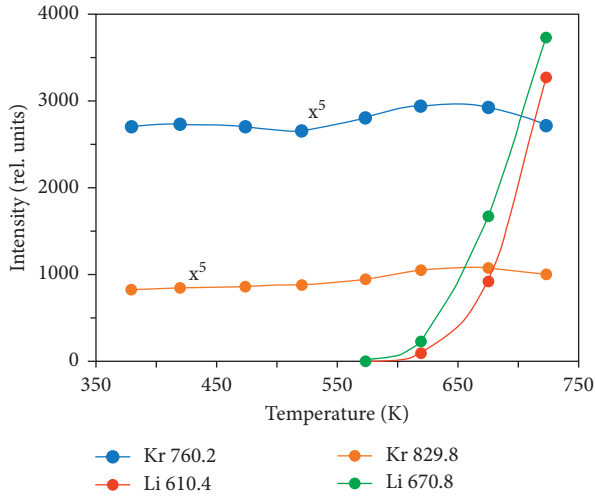


FIGURE 6: Dependence of the intensity of krypton and lithium lines in Ar-Kr mixture on the temperature of the lithium layer, $F = 10^{14}$ n/cm²s. The intensity of krypton lines increased by 5 times.

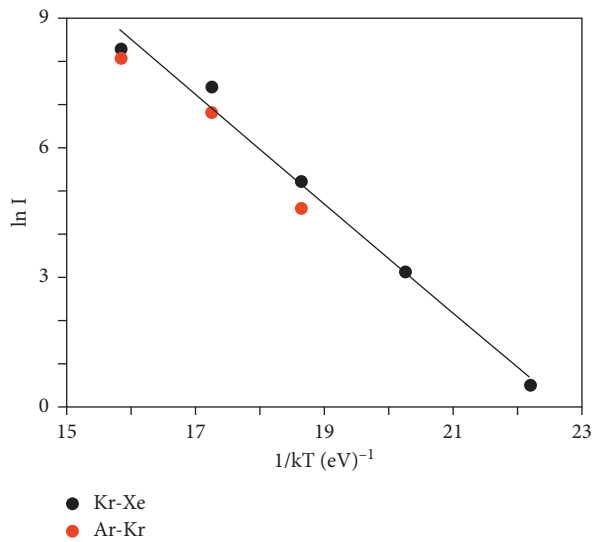


FIGURE 7: Dependence of 610.4 nm lithium line intensity in Kr-Xe and Ar-Kr mixtures on the inverse temperature of lithium layer.

decrease at temperatures above 670 K associated with radiation trapping at resonant transitions. A sharp increase in the intensity of the lithium line 610.4 nm with increasing temperature is satisfactorily described by the ratio (Figure 7):

$$I \sim \exp\left(-\frac{A}{kT}\right). \quad (13)$$

where k is the Boltzmann constant and A is activation energy of radiation process. From slope of the straight line in coordinates $(\ln I, 1/kT)$, the value $A \approx 1.3$ eV was obtained, which is in satisfactory agreement with the lithium evaporation energy of 1.63 eV (156.9 kJ/mol [18]), as well as the value obtained with single-component noble gases 1.61–1.67 eV [6].

Therefore, the appearance of alkali metal lines is associated with the evaporation of the release of α particles and

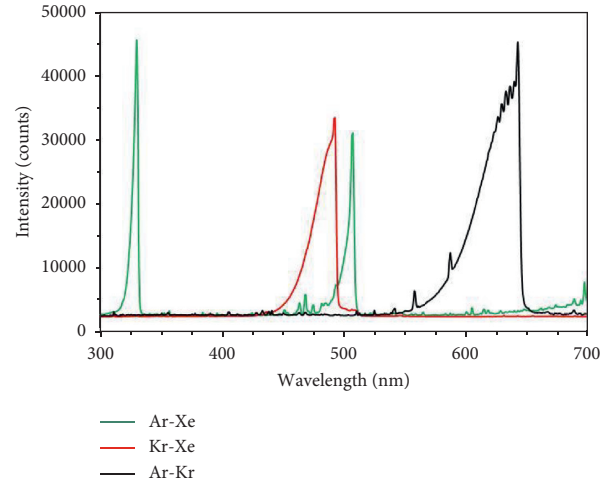


FIGURE 8: Emission spectra of gas mixtures at a temperature of 413 K Ar-Xe, Kr-Xe- $F = 10^{14}$ n/cm² s, Ar-Kr- $5 \cdot 10^{13}$ n/cm²s. Integration time: Ar-Xe-1 s Kr-Xe-100 ms, Ar-Kr-3 s.

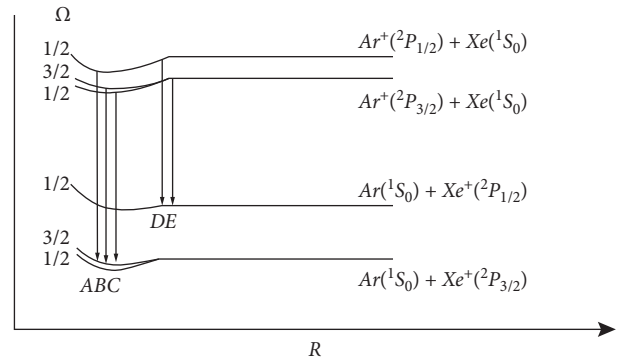


FIGURE 9: Illustrative diagram of potential curves of a $(\text{ArXe})^+$ molecule.

tritium nuclei from the deposited lithium layer, as well as in the case of single-component noble gases.

3.2. Emission of the Heteronuclear Ionic Molecules. The emission spectra of heteronuclear ion molecules are shown in Figure 8, a schematic representation of potential curves of the molecule $(\text{ArXe})^+$ is shown in Figure 9.

Maxima of bands of heteronuclear ion molecules are located at wavelengths 329 and 506 nm (Ar-Xe, bands A and D), 491 nm (Kr-Xe, band A), and 642 nm (Ar-Kr, band A). Similar bands were previously observed at excitation by an electron beam of Ar-Xe, Ar-Kr, and Kr-Xe mixtures [8, 9], as well as at excitation of mixtures by isotope decay products [10, 11, 19]. Under ionizing pumping, in contrast to the case of excitation by an electric discharge [7, 20], only bands A and D were observed. This is due to a sharp difference in rate constants of processes for states of the atomic ion $M^+ (^2P_{1/2})$ and $M^+ (^2P_{3/2})$. For mixtures of Ar-Xe and Ar-Kr, the rate constants of processes competing with the formation of heteronuclear ion molecules are as follows:

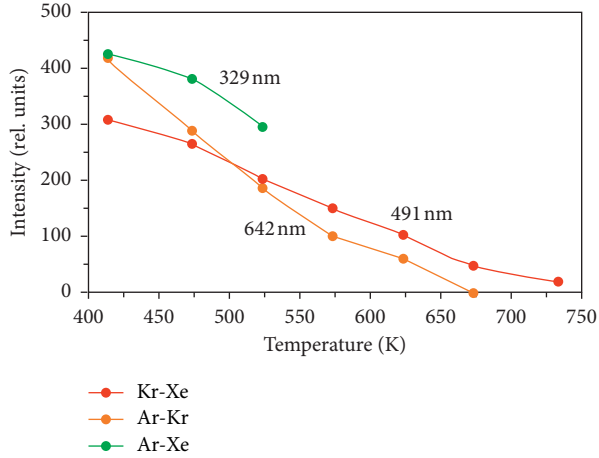


FIGURE 10: Dependence of the intensity of bands of heteronuclear ion molecules on the temperature of gas mixtures.

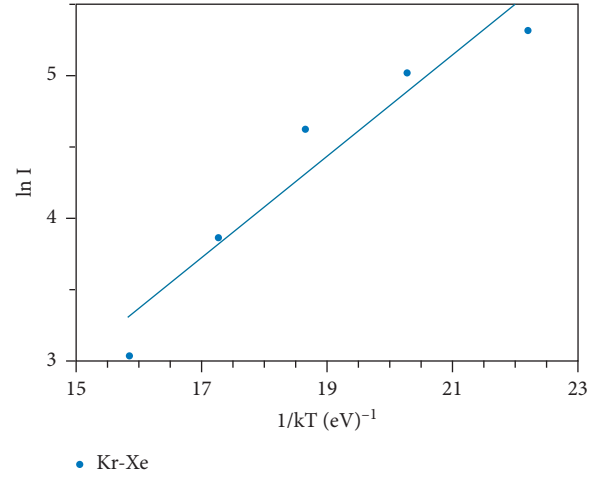
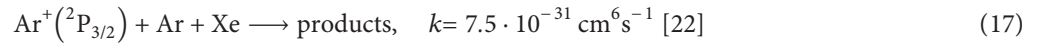
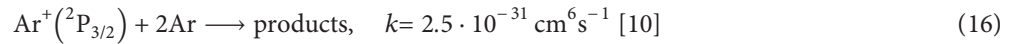
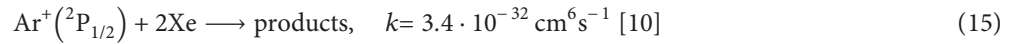
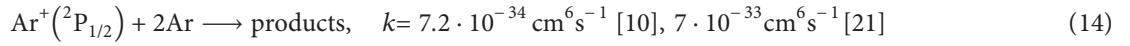


FIGURE 11: Dependence of the intensity of 491 nm band in Kr-Xe mixture on the reverse temperature of gas mixture.



The intensity of the B, C, and E bands should be much lower than the intensity of the A and D bands. The presence of the B, C, and E bands in [7, 20], apparently, is associated with the low pressure of the gas mixture in these works, 35 Pa [20] and 0.6–2 kPa [7]. At low pressure of the mixture, the difference in rate constants of processes involving ions $\text{M}^+(\text{}^2\text{P}_{1/2})$ и $\text{M}^+(\text{}^2\text{P}_{3/2})$ is less pronounced.

The temperature dependence of the radiation intensity in bands of heteronuclear ion molecules is shown in Figure 10.

The radiation in the 642 nm band of the Ar-Kr mixture disappears at a temperature of 670 K, and the radiation intensity of the Kr-Xe mixture (by a maximum of 491 nm) monotonically decreases to a maximum temperature of 730 K. Based on the relation for the temperature dependence of radiation,

$$I \sim \exp\left(-\frac{Q}{kT}\right). \quad (18)$$

An estimate was made of the value of Q —the dissociation energy of a heteronuclear molecule in a state with $\Omega = (1/2)$. For the $\text{Kr}^+(\text{}^2\text{P}_{1/2})\text{Xe}$ molecule, the value $Q = 0.36 \pm 0.10$ eV was obtained (Figure 11).

4. Conclusions

The radiation spectra of Kr-Xe, Ar-Kr, and Ar-Xe mixtures excited by the products of ${}^6\text{Li}(n, \alpha){}^3\text{H}$ nuclear reaction in the core of a stationary nuclear reactor at a thermal neutron flux density of up to 10^{14} n/cm² s were studied. In atomic

spectra, lines of $2p$ - $1s$ transitions of a heavier gas predominate; molecular spectra are represented by strong bands of heteronuclear ion molecules. With increasing temperature of the lithium layer, lines of lithium appear, as well as sodium and potassium, contained in lithium in the form of impurities. Also, as in the case of single-component noble gases, the appearance of alkali metal lines is associated with evaporation during the passage of nuclear particles from the lithium layer. The appearance of strong lines of lithium and sodium does not affect the intensity of atomic lines of noble gases, bands of heteronuclear ion molecules. Apparently, the main channel for the excitation of alkali metal atoms is the transfer of excitation in the process of collision of noble gas atoms in the $1s$ states (two metastable and two resonance, the radiation from which is trapped) with alkali metal atoms and subsequent ion-molecular processes in the plasma. The results obtained are interesting for the development of methods for removing energy from a nuclear reactor in the form of optical radiation. Selective deactivation of the $1s$ -levels of noble gases indicates the possibility of creating a laser at $2p$ - $1s$ transitions of noble gas atoms with the deactivation of the lower laser level by lithium atoms when excited by the products of the ${}^6\text{Li}(n, \alpha){}^3\text{H}$ nuclear reaction or electron beam.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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