

Optical Radiation from the Sputtered Species under Excitation of Ternary Mixtures of Noble Gases by the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ Nuclear Reaction Products

K. Samarkhanov^{1*}, M. Khasenov^{1,2}, E. Batyrbekov³, Yu. Gordienko¹,
Yu. Baklanova¹, I. Kenzhina⁴, Ye. Tulubayev¹, I. Karambayeva¹

¹Institute of Atomic Energy Branch of the National Nuclear Center of the Republic of Kazakhstan, 10 Beybit Atom str., Kurchatov, Kazakhstan

²School of Sciences and Humanities, Nazarbayev University, 53 Kabanbay Batyr Ave., Nur-Sultan, Kazakhstan

³National Nuclear Center of the Republic of Kazakhstan, 2 Beybit Atom str., Kurchatov, Kazakhstan

⁴al-Farabi Kazakh National University, 71 al-Farabi Ave., Almaty, Kazakhstan

Article info

Received:

1 February 2021

Received in revised form:

5 April 2021

Accepted:

22 May 2021

Keywords:

Nuclear reaction

Sputtering

Noble gases

Alkali metals

Level population

Abstract

The present paper examines the luminescence of ternary Ar-Kr-Xe and Ne-Ar-Kr mixtures of noble gases in the spectral range from 300 to 970 nm, excited by the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ nuclear reaction products in the core of a nuclear reactor. A thin layer of lithium applied on the walls of the experimental device, stabilized in the matrix of the capillary-porous structure, serves as a source of gas excitation. During in-pile tests, conducted at the IVG.1M research reactor, thermal neutrons interact via the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ reaction, and the emergent alpha particles with a kinetic energy of 2.05 MeV and tritium ions with a kinetic energy of 2.73 MeV excite gaseous medium. The study was carried out in a wide temperature range. The temperature dependence of the intensity of the emission of the atoms of noble gases and alkali metals, heteronuclear ionic molecules of noble gases were studied. The obtained values of the activation energy of the emission process 1.58 eV for lithium and 0.72 eV for potassium agree well with the known values of evaporation energy. Excitation of alkali metals atoms occurs consequently of the Penning process of alkali metals atoms on noble gas atoms in the 1s-states and further ion-molecular reactions.

1. Introduction

The direct conversion of the energy of nuclear reactions into optical radiation seems to be an important technical problem with great practical applications [1]. The solving of this problem will allow creation of highly efficient energy sources of coherent and non-coherent optical radiation [2]. The single-stage character of nuclear particle energy conversion into optical irradiation that bypasses intermediate stages of thermal, mechanical, and electrical energy, possesses higher efficiency, and the devices on its basis have low weight characteristics in comparison with similar devices.

In experiments to study the optical radiation of nuclear-excited plasma conducted at nuclear reactors, direct excitation of gaseous medium is usually carried out not by neutron radiation, but by using the products of exothermic nuclear reactions occurring during the interaction of thermal neutrons with ${}^3\text{He}$, ${}^{235}\text{U}$, ${}^{10}\text{B}$, ${}^6\text{Li}$ nuclei. There are two main methods of excitation: gaseous isotope or its compound (${}^3\text{He}$, ${}^{235}\text{UF}_6$) are part of the gaseous medium; the inner surface of the chamber is coated with a thin layer of an isotope or its compound.

Fission fragments of uranium-235, products of a nuclear reaction ${}^{10}\text{B}(n,\alpha){}^7\text{Li}$, were used as surface sources of charged particles. The relatively large mean free path of tritium nuclei in lithium and gaseous medium makes it possible to excite large volumes of gases and provide a larger amount of

*Corresponding author. E-mail: samarkhanov@nnc.kz

power deposited in the gas in comparison with the reaction products with ^{10}B . Our works using lithium as a surface source of excitation of gaseous medium in the core of a nuclear reactor, instead of the previously widely used uranium fission fragments, products of nuclear reactions $^3\text{He}(n,p)^3\text{H}$ and $^{10}\text{B}(n,\alpha)^7\text{Li}$ were aimed to find and study of the gaseous medium with high efficiency of converting the energy of nuclear reactions into optical radiation.

The temperature of the lithium cell housing during the previous reactor experiments with gas mixtures did not exceed $\sim 310\text{ K}$ [3, 4]. However, to reveal and define the patterns of emitting states formation in mixtures of noble gases with lithium vapor, a temperature range above the lithium melting point is required. The study of the influence of gas temperature on the kinetics of active medium population is a problem whose solution is necessary to understand and clarify the physical processes that occur during the creation of population inversion levels in these medium and to obtain the constants of elementary processes that are absent in the literature. In further reactor experiments, increasing the temperature of the gaseous medium up to $\sim 500\text{ K}$ led to the appearance in the emission spectra of lines of alkali metals [5, 6]. Not only lithium but also sodium and potassium lines contained as impurities in lithium are emitted. The appearance of these lines is associated with the evaporation of lithium under the emission of α -particles and tritium nuclei from the applied layer [5]. The release of excited atoms from the surface layer cannot provide emission from the gas volume; the near-surface layer will be emitted.

The luminescence spectra during lithium sputtering into a vacuum at room temperature of a target, irradiated with Xe^+ ions were studied in [7]. The processes leading to light emission during metal sputtering into a gaseous medium [5, 8] differ from the sputtering processes in the vacuum [9]. Cadmium and zinc sputtering in the gaseous medium during irradiation by the products of nuclear reactions $^3\text{He}(n,p)^3\text{H}$, α -particles of ^{238}Pu decay and an electron beam were studied in [8, 10, 11].

In [12], the possibility of simultaneous radiation at transitions of atoms of noble gases and alkali metals, as well as transitions of heteronuclear ion molecules of paired mixtures of noble gases [13, 14], excited by the $^6\text{Li}(n,\alpha)^3\text{H}$ nuclear reaction products were studied.

Experiments with Ar-Kr-Xe and Ne-Ar-Kr mixtures were a continuation of the series of reactor

experiments with multicomponent noble gases. The results are of interest from the point of view of widening the spectral range and increasing the efficiency of conversion of nuclear energy into light.

2. Experimental part

Most of the experimental work related to the research of kinetic energy conversion of nuclear reaction products into optical radiation was performed on pulsed nuclear reactors. At the same time, experimental work on the study of spectral-luminescent characteristics of nuclear-excited plasma was successfully carried out at nuclear reactors providing the possibility of long-term operation at constant power – in a stationary mode. Despite the relatively small neutron fluxes in stationary nuclear reactors, the presence of a continuous excitation source allows studying the optical radiation of a nuclear-excited plasma in a wide range of parameters (temperature, the pressure of the studied gaseous mixtures) in sufficient detail.

The studies were carried out at the IVG.1M research reactor [15]. To conduct reactor experiments, at various times, experimental devices with lithium as a surface source of charged particles were developed and successfully tested.

The results of the operation of the experimental device (Fig. 1a) with a rotating mirror and the external placement of the collimating lens led to significant light losses. Therefore, it was decided to place the collimating lens directly inside into an experimental device in follow up reactor experiments (Fig. 1b, c). This modification reduced the volume of the experimental device that makes it possible to decrease the effect of convective fluxes in a gas medium.

The design of the experimental device (Fig. 1c) markedly (3.5 times) increased the volume of the experimental cell, as well as the surface area of the lithium (1.3 times). The presence of a heater along the entire length of an experimental device and thermocouples, located at 4 points along the height of the outer surface made it possible to control the temperature gradient across the volume of the device during the experiments. The main difference in the design of this device from previous ones was the use of a capillary-porous structure (CPS) [16], which made it possible to stabilize lithium in a liquid state on the walls of an experimental cell during reactor experiments. A detailed description of the technique for applying a layer of lithium to the inner surface of the cell is presented

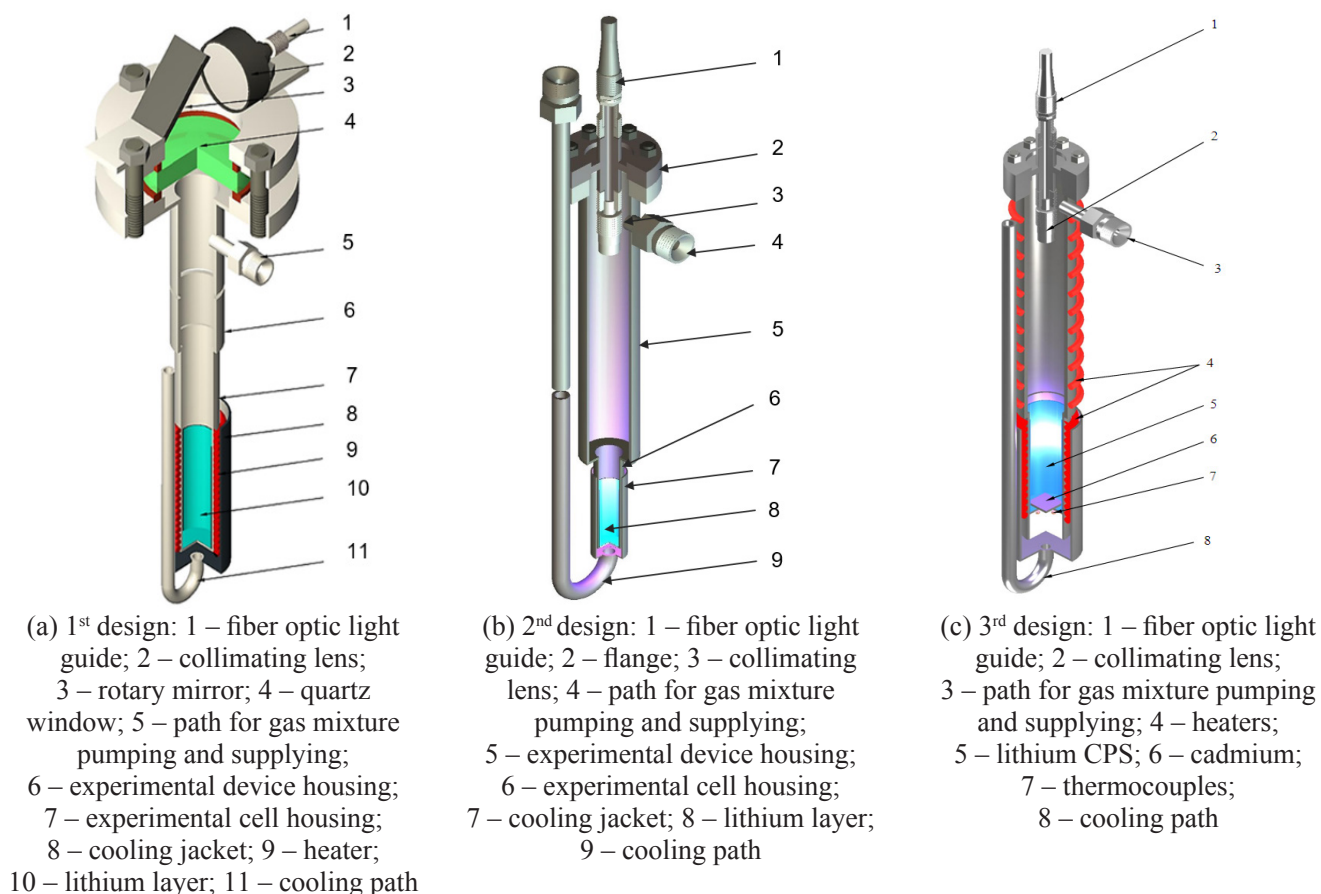


Fig. 1. Modifications of experimental device designs.

in [12]. In all experiments, LE-1 grade lithium (with a ${}^6\text{Li}$ isotope enrichment of $\sim 7.5\%$), with impurity content of less than 0.1% was used. The average thickness of the applied lithium layer (0.1 mm) was determined by calculation, based on the amount of loaded lithium, as well as the area of the inner surface of the CPS matrix.

A metallic cadmium plate weighing 1.42 g was placed on the bottom of the cell. It was assumed that it would be possible to compare the two mechanisms of optical radiation formation during thermal evaporation and during sputtering of the layer (respectively, cadmium and lithium). However, the radiation intensity of the brightest line of the cadmium atom (508.6 nm) did not exceed 150 rel. units . It is possible that the low intensity of the cadmium lines is associated with the formation of a cadmium-lithium compound.

The general procedure of the reactor experiments was conducted according to the early developed methodology [6]. The measurements were carried out at constant thermal power (6 MW) of the IVG.1M research reactor with a maximal thermal neutron flux density of up to $1.4 \cdot 10^{14}\text{ n/cm}^2\text{s}$. In this research the middle of the lithium layer was located 150 mm above the center of the reactor

core, which corresponded to the average thermal neutron flux density (Φ) $5 \cdot 10^{13}\text{ n/cm}^2\text{s}$. The frequency of nuclear reactions of lithium-6 with thermal neutrons is determined by the ratio:

$$S = \sigma\Phi N \quad (1)$$

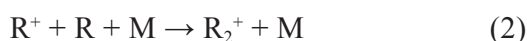
where $\sigma = 9.45 \cdot 10^{-22}\text{ cm}^2$ – ${}^6\text{Li}(n,\alpha){}^3\text{H}$ nuclear reaction cross section, N – amount of lithium-6 nuclei in the layer.

Occurring non-coherent optical radiation in the experimental device, as a result of excitation of the gaseous mixture by the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ nuclear reaction products outputted from the reactor core using a 10 m long fiber-optic cable. Remote recording of luminescence spectra was carried out by an operator using a QE65Pro-abs optical spectrometer (Ocean Optics). The luminescence spectra were recorded in the range of $300\text{--}970\text{ nm}$ with spectral integration times of $0.01\text{--}10\text{ s}$, spectrometer resolution 1.5 nm . The spectra are given without correction for the spectral sensitivity of the setup. The number of pulses of the spectrometer at a constant radiation intensity was proportional to the integration time. Measurements in a wide range of integration times allowed us to determine the intensities of both weak and strong lines.

Before the reactor reached a power level, the experimental device was pumped out and heated at a temperature of about 410 K. Gas pumping was performed using magnet-discharge pumps. Gas mixtures were preliminarily prepared in a separate tank and supply into the device after the reactor reached a power level of 6 MW. After filling the gas mixture the device was overlapped by a valve on the cup of the reactor at a distance of 2 m from the device center. Control of gas supply into the volume of the device up to the set value of pressure was performed using vacuum and pressure sensors. The temperature of the cell and the housing of an experimental device were regulated by changing the heater power, and if necessary, by increasing the nitrogen flow rate with external blowing. A planned shutdown of the reactor was performed, after the luminescence spectra of the studied gaseous mixture have been recorded.

3. Results and discussion

Gaseous mixture Ar:Kr:Xe=70:20:10 has been studied; total gas pressure was equal to 93 kPa under initial temperature of 391 K. Figure 2 demonstrates emission spectra of the mixture under various temperatures. Lines of the 2p-1s-transitions (Paschen notations) of xenon predominate under initial temperature; from atoms there are also krypton lines and weak argon line at 750.4 nm. When α -particles and tritium nuclei pass through a gaseous medium, $\sim 2 \cdot 10^5$ electron-ions pairs are formed (R^+ , e, where R – noble gas atom). In the processes of conversion of atomic ions R^+ (M – the third particle):

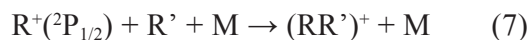


molecular ions R_2^+ are formed. R_2^+ ions are recharged on heavier gas atoms with lower ionization potential (finally on xenon atoms) and as a result, emission takes place mainly on the lines of 2p-1s-transitions of xenon atoms:



The part of ions R^+ in the mixture is involved in the formation of heteronuclear ionic molecules

of noble gases $(RR')^+$ (R' – atom of heavier gas) [13]. As a result of optical transitions from high ion levels, the ions remain in the states $R^+(^2P_{1/2})$ and $R^+(^2P_{3/2})$. In the processes:



molecules of heteronuclear ionic molecules $(RR')^+$ are formed. $(RR')^+$ molecule formation rate with the participation of the $R^+(^2P_{3/2})$ ions hundreds and thousands of times lower due to competitive processes [14]. Bands of molecules $(ArXe)^+$ (329 nm and 506 nm; they emit from general level), $(KrXe)^+$ (491 nm), $(ArKr)^+$ (642 nm) are shown in Fig. 2.

Figure 3 demonstrates the dependence of the intensity of xenon lines and bands of heteronuclear ion molecules on the temperature of a cell. Here and further, 1 relative unit of intensity corresponds to 1 thousand pulses of the spectrometer, normalized for the integration time of 3 s. The intensity of the molecular band is taken as the intensity at the maximum point, the contours of the molecular bands did not change with temperature changes.

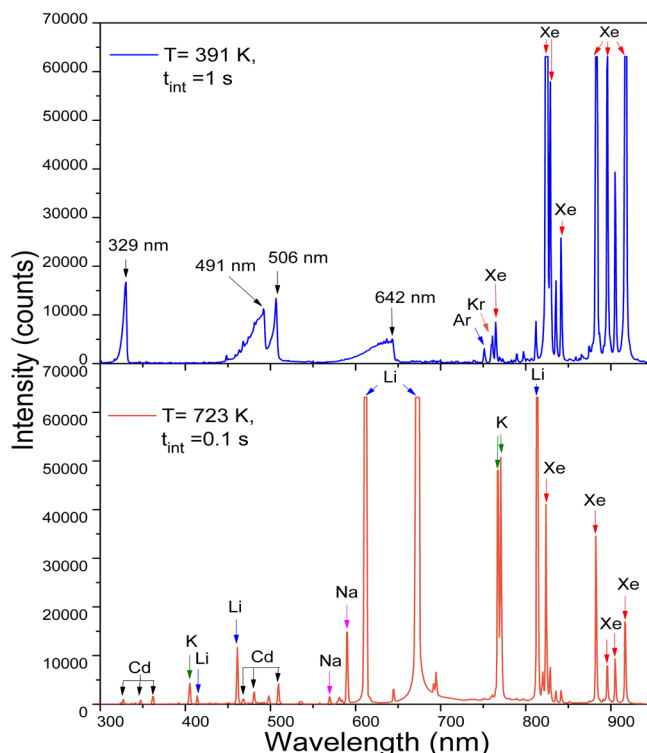


Fig. 2. Emission spectra of Ar-Kr-Xe mixture under an initial pressure of 93 kPa and temperatures of lithium layer 391 K and 723 K. An integration time of spectra is 1 s and 0.1 s, respectively. (Intensity of lines: xenon 823.2 nm – 332 000 counts, 881.9 nm – 180 000, 895.2 nm – 66 000, 916.3 nm – 163 000 at T = 391 K; lithium 610.4 nm – 368 000, 670.8 nm – 317 000, 812.6 nm – 396 000 at T = 723 K).

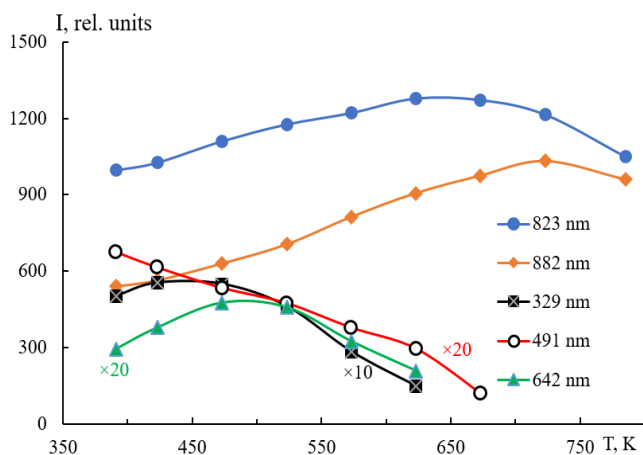


Fig. 3. Dependences of the intensities of xenon atom lines (823.2 nm, 881.9 nm) and bands of heteronuclear ion molecules $(\text{ArXe})^+$ (329 nm), $(\text{KrXe})^+$ (491 nm), $(\text{ArKr})^+$ (642 nm) in the mixture Ar-Kr-Xe from the temperature in the experimental device.

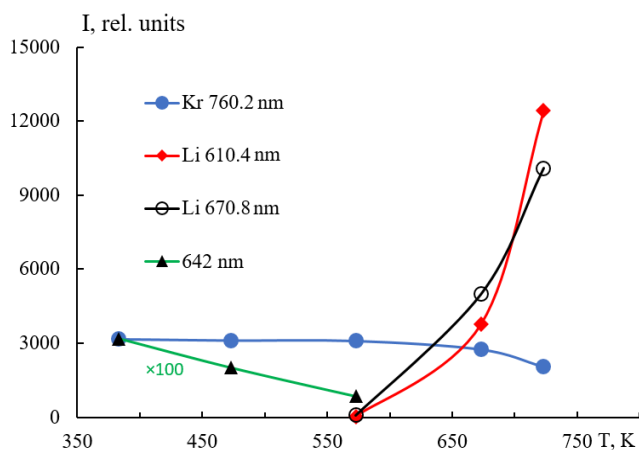


Fig. 4. Dependences of the intensities of krypton and lithium lines as well as a band $(\text{ArKr})^+$ on the temperature of lithium layer in Ne-Ar-Kr mixture.

The intensity of xenon lines grows monotonically with increasing temperature up to 700 K. The increase in the intensity of xenon lines is probably due to the dissociation of molecular ions $(\text{ArXe})^+$ and $(\text{KrXe})^+$, involving formed Xe^+ ions in plasma-chemical processes in the mixture.

For comparison, a Ne:Ar:Kr=70:20:10 gas mixture was studied, the total gas pressure was also 93 kPa at an initial temperature of 383 K. In this case, there is a band of only one heteronuclear ionic molecule $(\text{ArKr})^+$ with a maximum at 642 nm, the bands of $(\text{NeAr})^+$ and $(\text{NeKr})^+$ are located in the UV-region [13], which is beyond the spectral sensitivity of the setup. Of atomic lines, the lines of the 2p-1s-transitions of krypton predominate under initial temperature, there is also a weak argon line

at 750.4 nm. In the Ne-Ar-Kr mixture the intensity of the brightest krypton line remained constant up to the temperature reaches 670 K (Fig. 4). Apparently, the dissociation of the molecule $(\text{ArKr})^+$ did not significantly affect the population of krypton levels; band 642 nm is weak in comparison with $(\text{ArXe})^+$ and $(\text{KrXe})^+$ in Ar-Kr-Xe.

When the temperature rises up to 520 K, lines of alkali metals appear in the emission spectra of gas mixtures: lithium, sodium and, potassium. Alkali metal atoms have a similar level structure. Figure 5 shows the diagram of some lithium levels and optical transitions, observed in the experiment.

The dependence of the intensity of alkali metal lines on the temperature of the lithium layer in the mixture Ar-Kr-Xe is shown in Fig. 6.

Emission in the resonance lines of lithium at 670.8 nm (two lines are not resolved by the spectrometer) was “trapped” and subject to self-absorption. Emission in the resonance lines of sodium at 589 nm and 589.6 nm (3s-3p transition, these lines are also not resolved) was also trapped, and sodium lines are also observed at 568.3 nm (3p-4d) and 819.5 nm (3p-3d) in the spectra. The 766.5 nm and 769.9 nm (4s-4p) and 404.4 nm and 404.7 nm (4s-5p) resonance lines are also observed in the potassium spectra. Sodium and potassium are found in lithium as impurities with chemical compositions of 0.04 and 0.005%, respectively. The percentage of sodium and potassium in lithium may change during eight reactor cycles measurements of 3 h each.

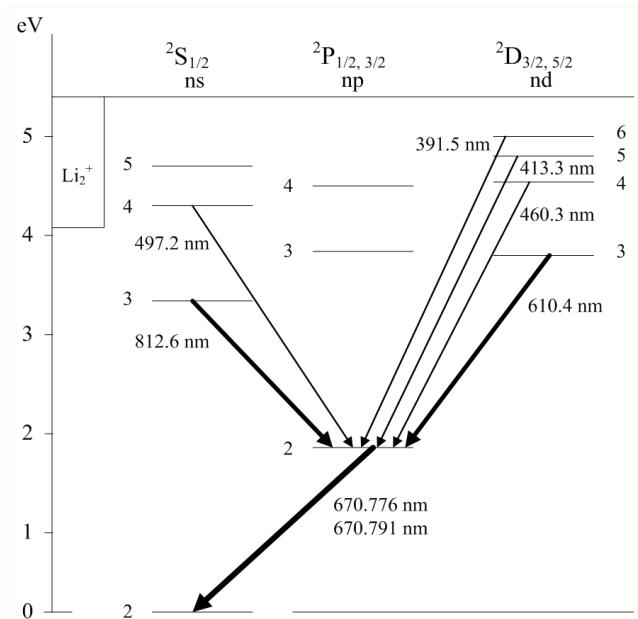


Fig. 5. Diagram of some lithium levels. Rounded values of the wavelengths of lithium atom lines are indicated except for resonant ones, without dividing them into separate multiplets.

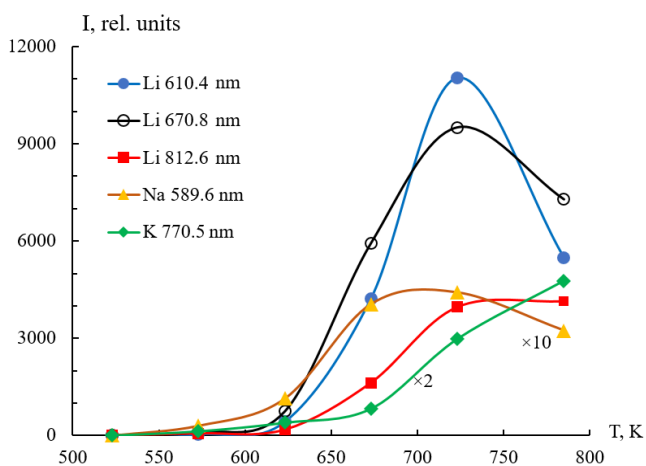


Fig. 6. Dependences of the intensities of alkali metal lines on the temperature of lithium layer in the mixture Ar-Kr-Xe.

Trapping of radiation occurs as a result of multiple reemission and reabsorption of resonance photons in an optically dense medium. Radiation trapping leads to a decrease in the intensity of the resonant lines of lithium and sodium at temperatures above 720 K in the mixture Ar-Kr-Xe. No visible signs of radiation trapping on the lithium line in the mixture Ne-Ar-Kr (see Fig. 4) is apparently associated with a more effective quenching of the resonant level of lithium (2p) by neon compared to other noble gases. In unary gases, radiation trapping on the resonance line of lithium was noticeable in krypton and xenon and was absent in neon and argon. Moreover, in krypton, radiation on the resonance line of lithium is trapped so strongly that the radiation is also trapped on 3d-2p-transition (610.4 nm). In binary mixtures, radiation trapping on the lithium resonance line was observed in the Kr-Xe mixture and was absent in the Ar-Kr mixture. Apparently, lighter neon and argon more effectively deactivate the resonance level of lithium.

Since the saturated vapor pressure of lithium is low ($1.0 \cdot 10^{-6}$ Pa under 500 K and 0.23 Pa under 750 K [17]), the process of radiation cannot be associated with ordinary thermal evaporation of lithium [6]. Figure 7 shows the temperature dependence of lithium and potassium luminescence intensity in $(1/kT, \ln I)$ coordinates, where k is Boltzmann constant.

Rapid growth in luminescence intensity is well approximated by the ratio:

$$I \sim \exp\left(-\frac{A}{kT}\right) \quad (8)$$

where A – activation energy of this process. For the 610.4 nm lithium line, the value of $A=1.58 \pm 0.15$ eV was obtained. This value coincides well with the evaporation (sublimation) energy of lithium equal to 1.63 eV (156.9 kJ/mol [18]) and differ sharply from the activation energy for self-diffusion in lithium (55.2 kJ/mol [19]). The measured value of A is well consistent the obtained value for the case with unary gases $A=1.61-1.67$ eV [6]. A noticeable disarrangement with the value for binary gases $A=1.3$ [12] apparently, is within the margin of error (10–15%). The main margin of error is associated with the reproducibility of the lithium spectrum, temperature unevenness along the height of the lithium layer and the choice of the linear part on the graph. For potassium line 769.9 nm $A=0.72 \pm 0.08$ eV, that also agrees well with the value of potassium evaporation energy 0.79 eV (76.6 kJ/mol [19]).

Thus, the emission of alkali metal atoms when lithium is sputtering into a gas medium by the products of nuclear reaction is not associated with self-diffusion of excited atoms in lithium. In [8], the sharp temperature dependence of the intensity of the emission of cadmium atoms and ions upon irradiation of cadmium with α -particles is associated with self-diffusion in cadmium. Apparently, in the region of gas excitation, a density of metal vapor is formed that differs from the density of the saturated vapor. It can be assumed that a significant proportion of the metal in lithium is sputtered not only in the form of microdroplets [8, 10], but also in the form of separate neutral atoms [20]. The direct contribution of released ions and excited atoms to the radiation is insignificant, as evidenced by the absence of noticeable optical radiation in a vacuum at a lithium layer temperature of 623 K.

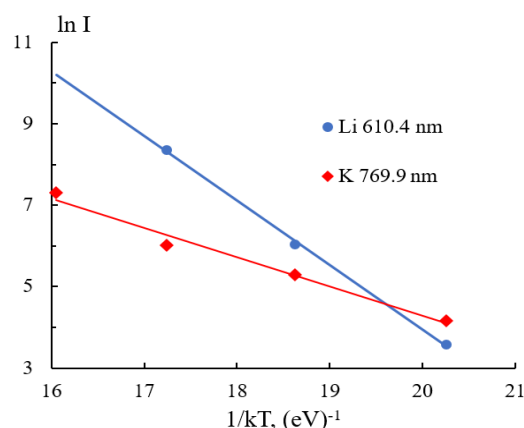


Fig. 7. Dependence of the intensities of lithium and potassium lines on the inverse temperature of lithium layer in the mixture Ar-Kr-Xe.

The intensity of lines of 2p-1s- transitions of noble gases increases with increasing temperature (see Fig. 3) or practically does not change (Fig. 4). There is no bend in the curves of the dependence of lines intensity of 2p-1s transitions of noble gas atoms on the temperature of the excitation region with a sharp increase in the intensity of alkali metal lines. The population of 2p-levels of lithium has almost no effect on the population of 2p-levels of noble gas atoms, including cascade transitions from higher levels. Thus, the main channel of the excitation transmission to lithium atoms is the Penning process on atoms of noble gases in 1s-states with the formation of atomic lithium ions:



Lithium atomic ions in processes of conversion (similar to process 4 for noble gas ions) form molecular ions Li_2^+ . Dissociative recombination of molecular lithium ions with electrons leads to the appearance of lines of lithium atoms in the spectra. Similar processes occur with sodium and potassium vapor.

4. Conclusion

Emission spectra of ternary Ar-Kr-Xe and Ne-Ar-Kr mixtures of noble gases, excited by the ${}^6Li(n,\alpha){}^3H$ nuclear reaction products in the core of the IVG.1M research reactor were studied. In atomic spectra, lines of transitions from 2p-levels of the heaviest gas atoms predominate up to temperatures of 600 K. Lines of alkali metal (Li, Na, K) atoms appear in the emission spectra of ternary gas mixtures when the temperature rises. Besides, as in the case of unary and binary noble gases, the appearance of alkali metal lines is caused by evaporation during the release of alpha particles and tritium nuclei from the lithium layer. However, the appearance of strong lines of alkali metals does not affect the intensity of atomic lines of noble gases. Apparently, the main channel for the excitation of alkali metal atoms is the transfer of excitation in the Penning process of collision of noble gas atoms in the metastable or resonance states with alkali metal atoms and further ion-molecular processes in the plasma.

In an Ar:Kr:Xe = 70:20:10 mixture, simultaneous emission occurs at the transitions of three heteronuclear ionic molecules: $(ArXe)^+$, $(ArKr)^+$ and $(KrXe)^+$. The intensity of the bands of heteronuclear ion molecules decreases noticeably at

temperature rises up to 650–700 K, when intense radiation is observed on the lines of alkali metals. The results obtained for Ar-Kr-Xe mixture are interesting for the development of methods for energy outputting from a nuclear reactor as of optical radiation.

Acknowledgments

This research is supported by the Ministry of Education and Science of the Republic of Kazakhstan (Grant No. AP08856017).

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