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Research Article

Method of Measuring the Efficiency of the Conversion of Nuclear Energy into Optical Energy

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A method of measuring the efficiency of converting nuclear energy into optical energy was developed based on correlations between intensities of the research line and the nitrogen second positive system in an Ar-N₂ mixture. In addition, the values of the coefficient of the conversion of nuclear energy into radiation at the lines of a Hg triplet in mixtures of Xe-Hg and Kr-Hg were determined. The values measured correspond to a selectiveness of pumping of 7^3 S₁ that was close to 1 ($\delta = 0.8 \pm 0.2$).

1. Introduction

The study of optical (laser and spontaneous) radiation of nuclear-excited plasmas is of interest for the development of a method to extract energy from nuclear reactors and to control and adjust nuclear reactors' parameters [1–3]. The essential parameters in the process of studying nuclear-excited plasma radiation are the values of the power of the nuclear reaction products deposited into gas and the efficiency of the conversion of nuclear energy into optical energy (η). The power of nuclear reaction products deposited into gas can be calculated for a specific geometry of the irradiating area or measured based on the gas overpressure after the pumping pulse [3]. The measurement of the parameter η is more complex, as one must measure the absolute intensity of the optical radiation from the area under consideration [4–6].

This paper focuses on developing a method for measuring the coefficients for converting nuclear energy into optical energy; our method involves radiation intensity comparisons in the mixture under study, where the radiation intensity was measured in a well-studied mixture. The studies were carried out with Polonium-210 α particle excitations.

2. Experimental Setup

The parameter η (the ratio of the optical power at the defined wavelength or molecular band to the power deposited into the gas) was determined by comparing the measured radiation intensities in the studied mixture with the intensity of the $C^3\Pi_u\text{-}B^3\Pi_g$ nitrogen band in an Ar + N_2 mixture. The gas pressure was selected such that the maximal range of α particles with energies of 5 MeV was similar for all the mixtures and the pumping power was comparable. A cylinder of diameter 25 mm and length 70 mm, carrying on its surface 18 sources containing ²¹⁰Po, was installed in a stainless steel chamber (Figure 1). The maximal range of α particles with energies of 5 MeV in Ar, Kr, and Xe under normal conditions is 37, 28, and 20 mm, respectively [7]. The activity of the α sources was 9.6 GBq, which corresponds to an average energy deposition W of approximately 10^{-4} W·cm⁻³ in 1.5 atm of Ar. Before we set up the sources, the chamber was heated and degassed under vacuum with a pressure of approximately 10^{-3} Pa. After the chamber was set up, the α sources were pumped down without heating for 2-3 weeks until the receipt of well-reproduced (up to 3–7% of intensity for various gases) luminescence spectra was verified. The gas pressure was measured by a standard mano-vacuum meter and VDG-1

TABLE 1: Rate constants of processes in an Ar-N ₂ mixture.

Number	Process	Constant of process	References
1	$\operatorname{Ar} + \alpha \rightarrow \operatorname{Ar}^* + \alpha$	$E_1 = 94 \mathrm{eV}$	[3]
2	$Ar + \alpha \rightarrow Ar^+ + \alpha + e$	$E_2 = 26.4 \mathrm{eV}$	[3]
3	$Ar^{+} + 2Ar \rightarrow Ar_{2}^{+} + Ar$	$k_3 = 2.1 \cdot 10^{-31} \mathrm{cm}^6 \mathrm{s}^{-1}$	[8]
4	$Ar^{+} + N_{2} \rightarrow N_{2}^{+} + Ar$	$k_4 = 1 \cdot 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	[12]
5	$Ar_2^+ + e \rightarrow Ar^* + Ar$	$k_5 = 6.9 \cdot 10^{-7} \mathrm{cm}^3 \mathrm{s}^{-1}$	[13]
6	$Ar^* + 2Ar \rightarrow Ar_2^* + Ar$	$k_6 = 8 \cdot 10^{-33} \mathrm{cm}^6 \mathrm{s}^{-1}$	[14]
7	$Ar_2^* \rightarrow hv + 2Ar$	$k_7 = 2 \cdot 10^8 \mathrm{s}^{-1}$	[8]
8	$Ar^* + N_2 \rightarrow N_2(B) + Ar$	$k_8 = 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	[15]
9	$\operatorname{Ar}^* + \operatorname{N}_2 \rightarrow \operatorname{N}_2(\operatorname{C}) + \operatorname{Ar}$	$k_9 = 3 \cdot 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	[15]
10	$Ar^* + N_2 \rightarrow N_2(E) + Ar$	$k_{10} = 5 \cdot 10^{-12} \mathrm{cm}^3 \mathrm{s}^{-1}$	[8]
11	$Ar_2^* + N_2 \rightarrow N_2(B) + 2Ar$	$k_{11} = 1 \cdot 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	[8]
12	$N_2(E) + Ar \rightarrow N_2(C) + Ar$	$k_{12} = 3 \cdot 10^{-12} \mathrm{cm}^3 \mathrm{s}^{-1}$	[8]
13	$N_2(C) \rightarrow hv + N_2(B)$	$k_{13} = 2.07 \cdot 10^7 \mathrm{s}^{-1}$	[15]
14	$N_2(C) + N_2 \rightarrow N_2(B) + N_2$	$k_{14} = 1.12 \cdot 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	[15]
15	$N_2(C) + Ar \rightarrow N_2(B) + Ar$	$k_{15} = 5.6 \cdot 10^{-13} \mathrm{cm}^3 \mathrm{s}^{-1}$	[16]
16	$N_2(B) \rightarrow hv + N_2(A)$	$k_{16} = 5.5 \cdot 10^4 \mathrm{s}^{-1}$	[15]
17	$N_2(B) + N_2 \rightarrow N_2(A) + N_2$	$k_{17} = 2 \cdot 10^{-12} \mathrm{cm}^3 \mathrm{s}^{-1}$	[8]
18	$N_2(B) + Ar \rightarrow N_2(A) + Ar$	$k_{18} = 1.4 \cdot 10^{-14} \mathrm{cm}^3 \mathrm{s}^{-1}$	[8]
19	$N_2(A) + N_2(A) \rightarrow N_2(E) + N_2$	$k_{19} = 1 \cdot 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	[8]
20	$N_2(A) + N_2(A) \rightarrow N_2(C) + N_2$	$k_{20} = 1 \cdot 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	[8]
21	$N_2(A) + N(A) \rightarrow N_2(B) + N_2$	$k_{21} = 8 \cdot 10^{-11} \mathrm{cm}^3 \mathrm{s}^{-1}$	[8]
22	$N_2(A) + N_2 \rightarrow N_2 + N_2,$	$k_{22} < 2.6 \cdot 10^{-18} \mathrm{cm}^3 \mathrm{s}^{-1}$	[17]
23	$N_2(A) + Ar \rightarrow N_2 + Ar$	$k_{23} < 2 \cdot 10^{-19} \mathrm{cm}^3 \mathrm{s}^{-1}$	[17]



FIGURE 1: The scheme of the chamber: 1: α -sources; 2: outgrowth with mercury; 3: windows; 4: cartridge.

vacuum meter, and the spent gases' purities were as follows: 99.992% for Ar and 99.999% for both Kr and Xe. The emission spectrum was analysed by a SPM-2 monochromator with a quartz prism and FEU-106 photomultiplier operating in the photon counting mode. The emission spectra were recorded under the maximal width of the monochromator slit; the forms of all the studied lines were similar to a triangle. In this case, the maximal value of the signal from the photomultiplier was proportional to the integral spectral intensity of the lines.

3. Efficiency of the Ar + N₂ Mixture's Luminescence

The efficiency of the Ar + N₂ mixture's luminescence was determined by calculation. A scheme of kinetic processes similar to the one described in [8] was used. The lifetime of Ar's metastable levels is approximately 50 s [9], and the radiation transferred from ${}^{3}P_{1}$ and ${}^{1}P_{1}$ into a normal state under atmospheric pressure was fully trapped. Therefore, all 4 levels of ${}^{3}P$ and ${}^{1}P$, as well as one level of Ar^{*}, were considered. Table 1 presents the processes relevant to the calculations and the proper rate coefficients.

Consider the transition $N_2(C^3\Pi_{u,v'=0}) \rightarrow N_2(B^3\Pi_{g,v''=i})$. The number of photons emitted in this transition is determined by the following formula:

$$I = A_{0,i} \left[N_2 \left(C^3 \Pi_{u,v'=0} \right) \right], \tag{1}$$

where the Einstein coefficients $A_{0,i}$ are presented in [10] and the density of nitrogen molecules on the $C^3 \Pi_{u,v'=0}$ level was calculated by considering processes 1–23. Moreover, the direct nitrogen excitation was insignificant, as our measurements show that the intensity of the nitrogen second positive system in N₂ (100 Torr) is approximately 140 times less than that in the Ar (1140 Torr) + N₂ (100 Torr) mixture. The intensity of the transitions from v' = 1 in the Ar + N₂ mixture was approximately 15 times less than the intensity of the transitions from v' = 0.



FIGURE 2: Dependence of radiation intensity (points) and luminescence efficiency (calculation by formula (2)) for $\lambda = 337.1$ nm on nitrogen pressure in Ar (1.5 Atm) + N₂ mixture.

The coefficient of the conversion of nuclear energy into optical energy can be determined by comparing the measured transition intensity (I_c) of the nitrogen molecules at the wavelength λ_c and the studied transition intensity (I_r) at the wavelength λ_r . A simple formula can be derived in the case of radioisotope pumping when processes 4, 22, and 23 can be ignored:

$$= \frac{A_{0i} \left[N_2 \right] \left[k_8 + k_9 + \left(k_{19} + k_{20} \right) \left(k_8 + k_9 \right) / \left(k_{19} + k_{20} + k_{21} \right) \right]}{\left(k_{13} + k_{14} \left[N_2 \right] + k_{15} \left[Ar \right] \right) \left\{ \left(k_8 + k_9 + k_{10} \right) \left[N_2 \right] + k_6 \left[Ar \right]^2 \right\}} \quad (2)$$
$$\cdot \frac{f \left(\lambda_c \right)}{f \left(\lambda_r \right)} \cdot \frac{I_r}{I_c} \cdot \frac{E_{21}}{E^*},$$

where E_{21} is the transition energy with wavelength λ_r , $E^* \approx 20.6 \text{ eV}$ is the energy consumed by the formation of one Ar^{*} atom in processes 1–5, and $f(\lambda)$ is the relative spectral sensitivity of the setup. To reduce the effect of the determination error of $f(\lambda)$ on the accuracy of the definition of η , N₂ molecular transitions closer to the wavelength of λ_r were selected.

The radiation intensity dependence measured in line 337.1 nm on the partial pressure of N₂ in an Ar + N₂ mixture is satisfactorily correlated with the dependence of η on the N₂ pressure within the range of 3–100 Torr (Figure 2).

4. Efficiency of the Luminescence of Gas Mixtures with Hg

In the mixtures of Xe or Kr with Hg, the 7^3S_1 level of the Hg atom is effectively populated by pumping with a rigid ionizer [11]. The main part of the energy (>98%) within 200–830 nm is emitted at triplet and resonance Hg lines (Figure 3). Because the harmful impurities that can result from radiation-chemical reactions are not accumulated in this mixture, it can be used as a scintillator for a nuclear-excited source of light. Thus, the values of the coefficient of



FIGURE 3: Emission spectrum of Kr (850 Torr) + Hg (1.5 mTorr) mixture: wavelengths (nm): 297 (1); 312.5 (2); 365 (3); 404.7 (4); 407.7 (5); 435.8 (6); 546.1 (7); 579 (8); lines intensity (rel. units): 3—14; 4—112; 5—10.7; 6—195; 7—149.

the nuclear energy conversion into optical energy in these mixtures have great significance.

The step of populating the 7^3S_1 level occurs in the processes of dissociative recombinations of molecular ions (Hg₂⁺, XeHg⁺, and KrHg⁺) with electrons [11]. Here, we describe the more essential processes in Xe + Hg plasma (where M is the third particle):

$$Xe + \alpha \longrightarrow Xe^+ + \alpha + e$$
 (3)

$$Xe^+ + Xe + M \longrightarrow Xe_2^+ + M$$
 (4)

$$Xe_2^+ + Hg \longrightarrow Hg^+ + 2Xe$$
 (5)

$$Hg^{+} + Xe + M \longrightarrow HgXe^{+} + M$$
 (6)

$$HgXe^{+} + Hg \longrightarrow Hg_{2}^{+} + Xe$$
 (7)

$$\mathrm{Hg_2}^{+} + e \longrightarrow \mathrm{Hg} \left(7^3 \mathrm{S}_1; 7^3 \mathrm{P} \right) + \mathrm{Hg}$$
 (8)

The $7^{3}S_{1}$ level is populated either in process (8) or by cascade transitions from the $7^{3}P_{0,1,2}$ levels.

In the recombination mechanism of the 7^3 S level, populating the dependence of the emission intensity of this level on Hg vapour's density is determined by the following formula [18]:

$$I = I_{\infty} \left(1 + \frac{\sqrt{\alpha S}}{k \,[\text{Hg}]} \right)^{-1},\tag{9}$$

where I_{∞} is the intensity under a high density of Hg atoms; k is the rate constant of charge exchanges of Xe₂⁺ (Kr₂⁺, KrXe⁺) ions at Hg atoms [19]; and the α coefficient of electron-ion recombinations is assumed to be identical for basic molecular ions ($\approx 10^{-6}$ cm³ s⁻¹).

By the above method, the ratio of the optical radiation's power to the power deposited into the gas (η) was determined for Hg triplet lines in mixtures of Xe + Hg and Kr + Hg. In terms of laser creation, the value of the 7³S₁ level pumping

selectivity (δ) is of interest. This value can be determined as the ratio of η to the quantum efficiency:

$$\eta_q = \frac{\tau}{W} \sum_{j=1}^{3} E_{1j} A_{1j},$$
(10)

where E_{1j} and A_{1j} are the energy and probability of a $7^{3}S_{1}-6^{3}P_{i}$ transition, respectively; τ is the lifetime of the $7^{3}S_{1}$ level; and *W* is the energy of an electron-ion pair formation in Xe or Kr. The mixtures' excitations were produced by α particles of ²¹⁰Po. The value η does not depend on the types of ionized particles that are produced [3], but it can depend on the value of the power deposited into the gas (q). Our analysis of the processes' kinetics in the studied mixtures showed that in the typical conditions for a stationary nuclear reactor ($q < 10 \text{ W} \cdot \text{cm}^{-3}$), the value of η under a sufficiently high pressure of Hg does not depend on the specific pumping power. Instead, the value of η is determined based on comparisons of the measured radiation intensities in the studied mixtures with the intensity of $C^{3}\Pi_{n}$ -B³ Π_{σ} in the Ar + N₂ mixture. η was measured under a chamber temperature of (295 ± 2) K, and the nitrogen pressure in the Ar + N₂ mixture varied from 10 to 100 Torr; the obtained values of η were averaged based on the measurements' results. The following values of η were obtained: 3.0% ($\lambda = 546$ nm), 3.6% (436 nm), and 1.5% (405 nm) were obtained for the mixture of Xe (760 Torr) + Hg (1.6 mTorr). The total value of η at Hg triplet lines comprised $(8.1 \pm 3.0)\%$ of the total. Under low Hg pressure, not all the Xe_2^+ ions can be recharged at Hg atoms. Thus, we recalculate η after considering dependency (9). At a sufficiently high Hg pressure ([Hg] $\gg \sqrt{\alpha S}/k$), the value of η is equal to (11 ± 5) %, which is sufficiently close to quantum efficiency (12%).

Regarding the mixture with Kr (850 Torr) + Hg (1.5 mTorr), the total value of η was equal to 11.2%. A recalculation under high Hg pressure yielded the value (13 ± 5)%, which is comparable with the quantum efficiency (11%). An η value exceeding the η_q value can be explained by experimental discrepancies and energy transfers from the Kr₂* level to the 7³S₁ level.

5. Conclusion

A measurement method was developed for the conversion of nuclear energy into optical energy, and this method is based on comparing the studied line intensity and nitrogen second positive system intensity in an Ar-N₂ mixture. The conversion efficiency level of nuclear energy was determined in radiated emissions on Hg triplet lines in Xe + Hg and Kr + Hg mixtures. The measured η values correspond to the selectivity of the 7³S₁ pumping level, which is close to 1 ($\delta = 0.8 \pm 0.2$). In the calculation model [20], the recombination flow is divided into even shares between the 7³P, 7³S₁, and 6³D levels. Because cascade transitions from 7³P populate the 7³S₁-level, this setup corresponds to a δ value of approximately 2/3. The method will be used to measure the coefficients of nuclear energy conversion into optical energy in gaseous mixtures in nuclear reactor cores.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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