

LASER FLUORESCENCE MONITORING OF THE GLOBAL RADIONUCLIDE IODINE-129

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A laser fluorescence system for monitoring the content of a long-living isotope of iodine-129 has been developed. The system uses a He-Ne laser as a light source. It provides the detection of this isotope in the process of technological refining of irradiated nuclear fuel at integrated radiochemical plant and in the atmosphere at the level of maximum permissible concentration.

1. The solution of the actual problem in ecologically safe functioning of enterprises with a nuclear fuel cycle requires new systems for monitoring the harmful leakages from these enterprises. This problem is of particular importance for monitoring of the global radionuclide iodine-129 which contaminates the atmosphere in the process of refining of the irradiated nuclear fuel since so far there are no highly sensitive means of detecting this isotope in real time.¹

At the same time it is known that in spite of the much smaller γ -activity of iodine-129 (whose half-life is 16 mln years) compared to a short-lived isotope of iodine-131 (this has made it feasible to detect iodine-131 by standard radiometric methods), its harmful effect on the man's organism is even stronger than that of iodine-131 being notorious after an accident at the Chernobyl' Atomic Power Station.²

We face two problems in monitoring of iodine-129. The first problem is to detect iodine with sensitivity of $\sim 10^{11}$ mol/cm³ directly in technological process of refining of irradiated nuclear fuel at integrated radiochemical plants. The second problem is ecological, i.e., monitoring of iodine-129 in the real atmosphere at a level of maximum permissible concentration being equal to $\sim 10^9$ mol/cm³. Substantially different composition and state of gaseous media and requirements for the sensitivity of iodine detection determine different approaches to the study and development of two different monitoring systems: one of them is intended for operation in a specific atmosphere and another – in the real atmosphere.

2. Methods of analytical laser spectroscopy, in particular, the method of laser-induced fluorescence, show significant promise for continuous monitoring of iodine contamination. Gas lasers, namely, argon ($\lambda = 515$ nm) and helium-neon ($\lambda = 633$ nm) lasers are most commonly used to produce iodine fluorescence. When detecting iodine-129 in a specific atmosphere, a sensitivity of $7.5 \cdot 10^{12}$ mol/cm³ was attained using an argon laser³ and a sensitivity of $5 \cdot 10^{12}$ mol/cm³ was obtained with a He-Ne laser.⁴ In the real atmosphere the highest sensitivity of $\sim 10^{11}$ mol/cm³ was also reached using a He-Ne laser.⁵

On this basis as well as considering high reliability, ease of operation, and sufficiently large resource of a He-Ne laser, we may conclude that it is this laser which must be used as a basis of laser-fluorescence systems for monitoring of iodine-129 in specific and real atmospheres. This problem is discussed in this paper.

2.1. We face the problem of selective detection of the given ¹²⁹I₂ isotope of iodine against the background of the ¹²⁷I₂

molecule in the real atmosphere.⁶ The contribution of each type of molecules to the recorded fluorescence signal can be separated by tuning the laser radiation frequency over resonance absorption lines of the aforementioned molecules.⁷ In our paper we used a He-Ne laser tunable in a longitudinal magnetic field. The range of frequency tuning was 5 GHz. Glass windows of a laser gas-discharge tube were orthogonal to the optical axis. The radiation components σ^+ and σ^- were separated using a quarter-wave plate and a polarizer, where σ^+ and σ^- are the clockwise and counterclockwise polarization components of laser radiation induced as a result of the imposition of a longitudinal magnetic field on a gas-discharge element of a laser.

The ¹²⁹I₂ fluorescence was produced in a measuring cell 3.5 cm in diameter and 41 cm long connected with a vacuum post for smooth feed of atmospheric air (Fig. 1). Pressure in the cell was adjusted using a standard vacuum gauge.

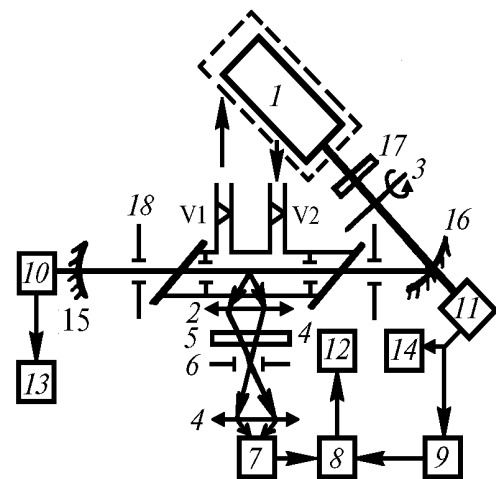


FIG. 1. Diagram showing the experimental configuration of the laser-fluorescence system: 1) laser system; 2) measuring cell; 3) laser beam modulator; 4) focusing lenses; 5) spectral light filter (the wave band is either 650–900 or 585–620 nm); 6 and 18) diaphragms; 7) photomultiplier; 8) synchronous detector; 9) amplifier of variable signal; 10 and 11) photodetectors; 12) IBM PC; 13 and 14) digital voltmeters; 15 and 16) external mirrors; 17) interference light filter (whose wave band is 590–640 nm).

The laser radiation was modulated with a low-frequency mechanical beam chopper. A valid signal was focused onto a photocathode of a photomultiplier with a system of fluorescence convergence which consists of two lenses and a diaphragm. A signal from the photomultiplier feeds into a synchronous detector with a 0.03 Hz bandwidth. The signal was recorded and processed on an IBM PC. A laser radiative power was measured in front of and behind the cell using photodetectors and was recorded by digital voltmeters. An absorbing spectral light filter with a 650–900 nm wave band was used to select a fluorescence signal against the scattered laser radiation background.

Spontaneous radiation of the laser gas–discharge tube falling within the light–filter wave band was eliminated with the help of an interference filter having a 590–640 nm wave band, which was placed in front of the cell. Moreover, the lower level of laser radiation scattered on parts of the measuring cell was provided by diaphragms of a laser beam set in and out of the cell and by blackening of the inner surfaces of the cell. To decrease dark current, the photocathode was cooled with liquid nitrogen.

A multipass system formed by external mirrors with the cell inserted between them was used to increase the sensitivity.

The setup sensitivity was $\sim 10^{-16}$ W and was limited by the fluctuations in the dark current of the photomultiplier.

Since the preparation of a reference cell with 100% content of $^{129}\text{I}_2$ was complex and costly, a cell filled with a mixture of molecular iodine isotopes in a $^{129}\text{I}_2$: $^{127}\text{I}_2$: $^{127}\text{I}_2$ = 0.74:0.24:0.02 ratio which is typical of irradiated nuclear fuel was used to estimate the detection sensitivity of iodine–129 in the real atmosphere. The pressure of saturated vapors of molecular iodine in the cell was determined by the ambient temperature and corresponded to iodine concentration of 10^{16} mol/cm³. Measurements were carried out in the cell under an air pressure of ~ 10 Torr close to optimal from a sensitivity standpoint. In this case the signal–to–noise ratio was $\sim 10^7$. As to the selectivity of separating a fluorescence signal of iodine–129 against iodine–127, the fluorescence intensity ratio $^{129}\text{I}_2$: $^{127}\text{I}_2$ was ~ 100 .

2.2. There are some peculiarities in detecting iodine–129 in a specific atmosphere (i.e., in a gaseous working medium on nitric–acid solution of the exhausted nuclear fuel). First, there is no need to use a frequency–tunable laser, since in a working mixture the content of iodine–129 is larger than that of iodine–127 by a factor of 5–6; moreover, it has the absorption coefficient which is larger by approximately one order of magnitude than that of $^{127}\text{I}_2$. Second, since the basic component of a specific atmosphere is nitrogen peroxide which strongly absorbs the He–Ne laser radiation, the problem arises of determining the iodine–129 concentration against the NO_2 fluorescence background. In connection with the fact that the He–Ne laser radiation induces practically only the Stokes region of the nitrogen peroxide fluorescence spectrum (650–900 nm), it is expedient to record iodine fluorescence in the region 585–620 nm (the anti–Stokes region).

The developed laser fluorescence system for detecting iodine–129 in a specific atmosphere was tested at the V.G. Khlopin Radium Institute. In the experiment a sample of the exhausted nuclear fuel as a fragment of a fuel element of a nuclear reactor of the atomic power station was dissolved. The element in use was held for a long time (≥ 14 years) and contained mainly two iodine isotopes: ^{129}I

and ^{127}I with a predominance of iodine–129 (84%). On solution of fuel in nitric acid, 95–97% of iodine was converted to a vapor–gas phase, with a predominance of gaseous iodine being in the form of molecular compounds $^{129}\text{I}_2$, $^{129}\text{I}^{127}\text{I}$, and $^{127}\text{I}_2$.

A linearly polarized multimode 25 mW laser was used to produce the fluorescence. The iodine fluorescence was recorded using interference light filters in the spectral range 585–620 nm. To decrease the level of laser radiation scattered on the cell parts, the cell windows were oriented at Brewster's angles. Moreover, the radiation scattered on microparticles of the working mixture under study was eliminated using an aerosol filter.

A small portion of a gas flow passing from a fuel–solution system to measuring cell, continuously monitored with a flowmeter, was sampled during the experiment (see Fig. 1). The pressure of gas mixture in the cell was 6–8 Torr. To decrease the measured iodine concentration, a gas purifying filter with the coefficient of iodine capture $\sim 10^2$, analogous to the filters used at radiochemical integrated plants, was placed at the cell input. A iodine trap was placed upon exiting the gas flow with the aim of independent testing of results obtained with the laser system. After the experiment the trap was used to determine the total quantity of iodine–129 separated on solution. The gas mixture passed through the setup was purified from harmful substances and entered the atmosphere.

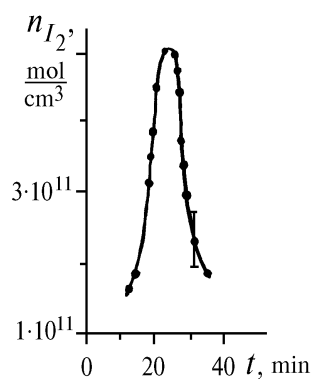


FIG. 2. Recorded fluorescence signal of iodine–129 vs. the time of fuel sample solution.

Depicted in Fig. 2 is a plot of a iodine fluorescence signal vs. the time of fuel–sample solution. It should be noted that the iodine–127 contribution to the observed signal did not exceed 1–2%. An instant of heated gas feed to the solvent setup was taken as a "zero" count of time. A cell with a reference mixture of molecular iodine isotopes corresponding to isotopic composition of iodine in a sample of irradiated fuel was used to calibrate the fluorescence signal obtained. The maximum iodine concentration in the solution was $(5.3 \pm 0.3) \cdot 10^{11}$ mol/cm³. It should be noted that the values of total content of iodine–129 measured by radiometric and laser–fluorescence methods differ by no more than 15%.

3. Thus the developed laser–fluorescence system is capable to monitor the content of iodine–129 both directly in a gas working medium after technological refining of the irradiated nuclear fuel and upon exiting the gas–purification systems at radiochemical integrated plants. Moreover, this system can also be used for ecological monitoring of iodine–129 in atmospheric air at a level of maximum permissible concentration.

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