Detection of Molecular Iodine Isotopes $^{127}$I, $^{129}$I, $^{131}$I in the Gases Using a Tunable Diode Laser

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Our previous papers report on using of diode lasers tunable in the range of 630-640 nm for selective on-line detecting of $^{129}$I and $^{127}$I molecular iodine isotopologues in gaseous media by laser-induced fluorescence (LIF) method [1]. Despite the achieved high sensitivity of $^{129}$I and $^{127}$I detecting, the question of iodine radionuclides monitoring during spent nuclear fuel (SNF) reprocessing is not completely settled. Thus, the problem of providing on-line control of short-lived isotope $^{131}$I with half-life of $\sim$8 days has not been solved. The paper reports on the developing of laser-induced fluorescence method for on-line selective measurement of $^{127}$I$_2$, $^{129}$I$_2$, $^{131}$I$_2$, $^{129}$I$^{127}$I, $^{127}$I$^{131}$I, $^{129}$I$^{131}$I isotopologues concentrations in gaseous media.

During SNF recycling in gaseous technological flow molecular iodine is formed as molecules of various isotope composition: $^{127}$I$_2$, $^{129}$I$_2$, $^{131}$I$_2$, $^{129}$I$^{127}$I, $^{127}$I$^{131}$I, $^{129}$I$^{131}$I. Concentrations of each of six iodine molecules can be measured at fluorescence exciting at six different wavelengths provided that absorption lines of iodine molecules do not overlap with each other, i.e. fluorescence of iodine molecule of given isotope composition only is excited at each wavelength.

This method cannot be put into practice, since reduced masses of $^{129}$I$_2$ and $^{127}$I$^{131}$I molecules are so close that isotope shift for them is almost the same and their absorption lines almost completely overlap with each other. That is why only four iodine molecules concentrations can be measured ($^{127}$I$_2$, $^{131}$I$_2$, $^{129}$I$^{127}$I, $^{127}$I$^{131}$I, $^{129}$I$^{131}$I), and concentrations of $^{129}$I$_2$ and $^{127}$I$^{131}$I molecules are to be calculated after that. Then the method of simultaneous measuring of six iodine molecules concentrations can be realized at fluorescence exciting at four wavelengths.

In order to choose fluorescence exciting wavelengths we carried out calculations of absorption and fluorescence spectra for molecular iodine isotopologues in spectral range of the used laser diode (632-637 nm). $^{127}$I$_2$, $^{131}$I$_2$, $^{129}$I$^{127}$I, $^{127}$I$^{131}$I isotope isotopologues have absorption lines, which almost non-overlap with absorption lines of all other isotopologues. Thus, $^{129}$I$^{127}$I isotope has two quite intensive absorption lines near 632.701 nm and one less intensive line near 632.08 nm, $^{129}$I$^{131}$I has two lines near 632.705 nm, $^{131}$I$_2$ has two lines near 632.097 nm and one less intensive line near 632.705 nm, $^{127}$I$_2$ has several lines near 632.077 nm.

Experimental procedure of the proposed method may be similar to the procedure used in paper [1]. In this procedure modulated laser radiation excites fluorescent emission in the experimental and reference cells at the given wavelengths, and then it is collected at an angle of 90° to the laser beam and focused on the photocathode of the photomultiplier tube. Then it is received by lock-in amplifier. The carried out research shows that boundary relations of iodine isotopologues concentrations, measured simultaneously, are at the level of $10^{-5}$-$10^{-6}$.

The conducted research shows that using diode laser tunable in the red spectral range as a source of fluorescence exciting is promising for highly-sensitive and high-precise on-line control of $^{127}$I, $^{129}$I, $^{131}$I iodine isotopes molecular forms in gaseous mixtures. Realization of the proposed method (in contrast to previously used methods) allows not only to control $^{131}$I isotope, but also to increase accuracy of $^{127}$I and $^{129}$I isotopes control.

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References