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Quick Detection of Uranium and Thorium Radioactive Materials in NORM Sample Using Laser Induced Plasma Spectroscopy Technique (LIPS)

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Abstract

Nuclear technology has been developed well in Indonesia because of its wide applications. The nuclear technology applications are not only for energy but also for health, agriculture and industry. The exploitation of nuclear technology should certainly consider to minimize the effects of nuclear radiation hazards both for workers and environment around the installations. It is conducted by monitoring, detection and measurement of such nuclear radiation within their operational area. Generally, it is carried out initially by nuclear detection devices, another method used for nuclear detection using Laser Induced Plasma Spectroscopy (LIPS). LIPS technique has fast qualitative and quantitative response, uses *in situ* sample analysis, and almost has no need sample preparation for its detection process. Nd-YAG laser Q-Switch 355 nm, 10 Hz, pulse duration of 5.50 ns, f = 100 mm, the energy 107 mJ was focused on Natural Occurring Radioactive Material (NORM) for its sample, under a surrounding air pressure of 1 Torr. It was demonstrated that good quality spectra were obtained with very low background for spectrum line of thorium and uranium .The results show that LIPS technique using optimum of 107 mJ laser energy qualitatively be able to identify the presence of radioactive elements, i.e. Uranium (U) and Thorium (Th) contained in the test sample. Predictive value of U is obtained 155 ppm with 11.30% error while its detection limit value is 7.89 ppm, and Th is obtained 124 ppm with 8% error while its detection limit value is 12.40 ppm. The experiment shows that LIPS technique is inherently very suitable and it is possible to be used as a measurement technique, analysis and identification of the presence of radioactive materials.

Keywords: LIPS; Laser Ablation; NORM; Uranium; Thorium; Nuclear.

Introduction

Recently publics are concern about radioactive material and their effect to human body or environment. In nature, there are various natural radionuclides. Quantitatively, the most abundant are potassium-40, U series, nuclides Th series nuclides, and these are called primordial

radionuclides, which are radionuclides that have existed in the Earth's crust since the formation of the universe. In addition, there are other natural radionuclides which are relatively small in number, namely various radionuclides such as Cosmo genic nuclides (nuclides resulting from interactions of cosmic radiation), which occur due to interactions between cosmic radiation and air, and nuclides of spontaneous decay products of nuclides can be split. The source of artificial radiation in the environment comes from nuclear testing, nuclear fuel cycles and other sources that are the result of activation. The activity of fallout radioactivity due to nuclear testing constitutes the largest part of artificial radiation activity. However, the activity of artificial radiation is relatively very small compared to natural radiation activity. Natural radioactive elements which are mobilized and then accumulated in end industry process are known as Naturally Occurring Radioactive Materials (NORM).

The level of danger and the nature of nuclear radiation that cannot be felt at all by the five human senses and can penetrate various types of materials. It is necessary to monitor, measure, analyze and identify the presence of such radioactive elements. The technique commonly used to analyze and identify the presence of radioactive elements is by using a nuclear detection device or nuclear radiation detector [1, 2].

In this experiment, an alternative method was carried out for the measurement, analysis and identification of radioactive elements in NORM sample from Ketapang west of Sumatra Island using LIPS techniques. Utilization of LIPS technology was chosen because LIPS is an in situ, qualitative and quantitative sample analysis technique that is fast and almost without sample preparation, and allows it to be done by remote sensing or remotely, and suitable for many kind of material [3 – 11].

Materials and Methods

Material used in these experiments are samples of NORM. Natural radioactive material comes from the Ketapang area West Kalimantan.

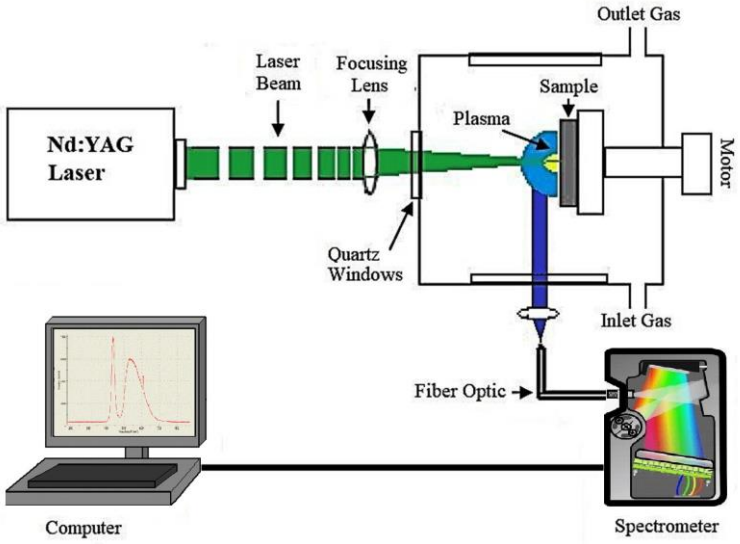


Fig. 1 Schematic diagram of the experimental set-up used in this study.

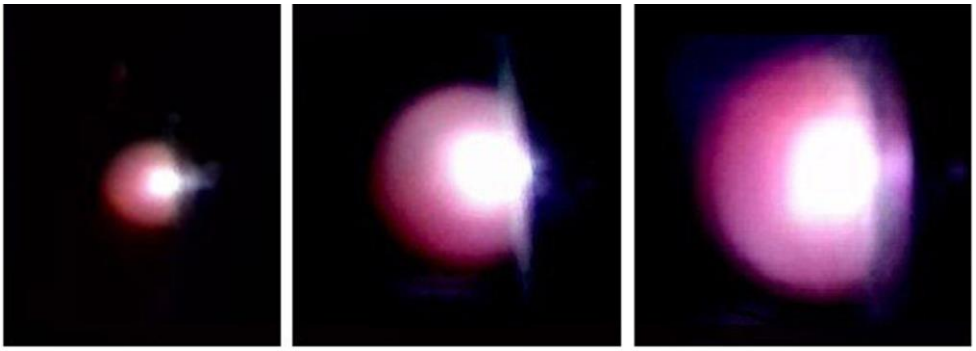
The equipment and Schematic diagram of the experimental set-up used in the study is shown in Fig. 1. The ND: YAG laser third harmonic 355 nm (Quantel-Q-Smart 850, 355 nm, pulse duration of 5.50 ns) is operated by Q-Switch 10 Hz with an energy output varies 5 mJ, 10 mJ, 18 mJ, 28 mJ, 55 mJ, 72 mJ, 107 mJ and 140 mJ used as a source of energy evaporation of atoms and molecules in the NORM sample. The NORM sample target was placed on inside the vacuum chamber with a size of 11 × 11 × 12.50 cm^3. The chamber could be evacuated with a vacuum pump and fill with a surround gas at the desired pressure. The gas flow through the chamber was regulated by a needle valve in the air line and a second valve in the pumping line. The chamber pressure was monitored by means of a digital Pirani meter (DIAVAC, PT-1DA) which is used to monitor the air pressure in the chamber, and quartz lens with a focal distance of 100 mm was used to focus the laser beam to the NORM surface sample. The condition of plasma observation was for low pressure with a fixed chamber pressure around 1 Torr and 760 Torr for the atmospheric plasma observation. The MayaPro2000 VIS-NIR spectrometer is used to detect plasma radiation and connected to an optical fiber with its entrance placed in front of the observation window of the vacuum chamber. In order to get the new position of sample during the laser irradiation, the sample was rotated so that the resulting data were the spatial averages of results obtained at different points in the sample.

(a) (b)

Fig. 2 Photograph of the sample used in this experiment. (a) original (b) after cutting.

Results and Discussion

The secondary plasma observed in these experiments is shown in Fig. 3 (a) hemispherical plasma related with previous experiments [12 – 14]. In these experiments we chose NORM as a sample and we examined the spectrum line of Th and U. The results show many lines of Th and U, but for more analysis we concern with Th II 530.46 nm and UI 395.15 nm. In order to obtain the suitable condition to take data, we choose low pressure 1 Torr and energy 107 mJ which shows that the emission intensity is high.



(a) (b) (c)

Fig. 3 Photograph of the plasma when an Nd-YAG laser 355 nm with different energy (a) 10 mJ (b) 72 mJ and (c) 107 mJ was focused on the NORM sample at 1 Torr.

In order to detect the present emission spectra of the both Th and U on NORM sample together, we prepare experiment with the variation energy of laser. The best condition found in energy is 107 mJ, because in this energy is not only detect the Th and U spectra line only but also the high emission intensity that can be present on both Th II 530.46 nm and U I 395.15 nm.

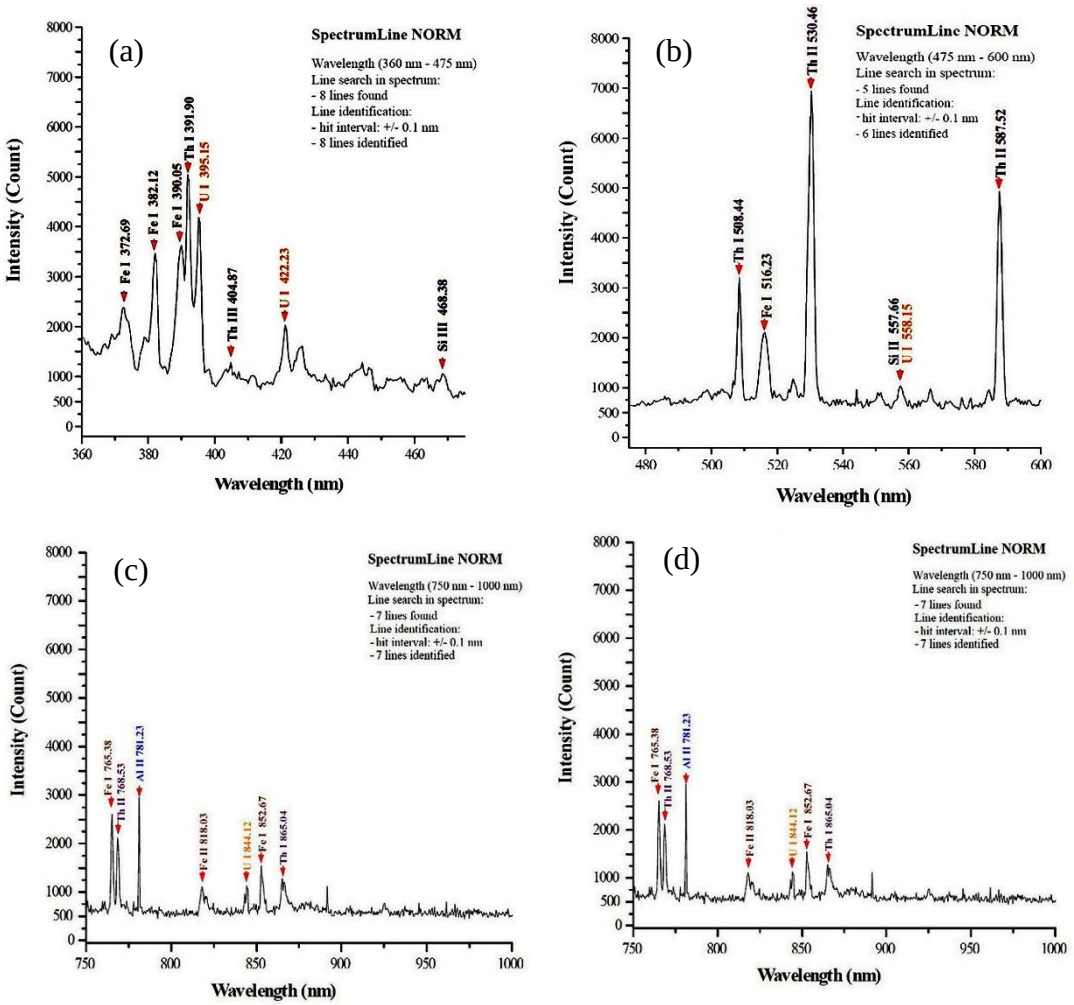


Fig. 4 Emission spectra of NORM sample in the wavelength ragion (a) 360 – 475, (b) 475 – 600 (c) 600 – 750, and (d) 750 – 1000 nm. These spectra were taken using 355 nm Nd-YAG laser and 1 Torr air.

Figure 4 shows the emission spectra of plasma generated using 355 nm Nd-YAG laser at 1 Torr air pressure and 107 mJ energy from NORM sample in the wavelength regions of (a) 300 – 475 nm, (b) 475 – 600 nm, (c) 600 – 750 nm, and (d) 750 – 1000 nm. In this paper it can be seen that many Th lines and U lines present in these results and these will be compared with Th and U lines from NIST table which as shown in Table 1.

Table 1 Competition of emission intensity of Th line and U line from NORM sample with NIST table.

No	Radioactive element	Wavelength (nm)	
		Experiment	NIST
1	Th I	391.90	391.90
2	U I	395.15	395.15
3	Th III	404.87	404.87
4	U I	422.23	422.23
5	Th I	508.44	508.44
6	Th II	530.46	530.46
7	U I	558.15	558.15
8	Th II	587.51	587.52
9	Th II	614.74	614.75
10	Th I	654.97	654.98
11	Th II	768.54	768.53
12	U I	844.12	844.12
13	Th I	865.03	865.04

The quantitative analysis is calculated for spectra line of Th II 530.46 nm and U I 395.15 nm using variable energy (5 mJ, 10 mJ. 18 mJ, 28 mJ, 55 mJ, 72 mJ and 107 mJ), because from energy of 5 mJ both Th and U are already detected. In order to make linear regression of Th II 530.46 nm and U I 395.15 nm, each energy of 3 times data collection was performed. The results obtained are for Th II 530.46 nm: $y = 59.0401x + 609.8395$ with $R^2 = 0.9094$ and for U I 395.15 nm: $y = 36.1532x + 244.2829$ with $R^2 = 0.8288$. By prediction quantitative calculation, the predicted value of U element concentration is 155 ppm with an error percentage of 11.30% and its limit detection value is 7.89 ppm. The predicted value of Th element concentration is 124 ppm with an error percentage of 8% and its limit detection value is 12.40 ppm.

Conclusion

The successful detection of the strong U and Th emission and other elements contained in NORM sample demonstrated using LIPS. In order to detection and analyze of U and Th in NORM sampel we used 107 mJ of optimum laser energy. The quantitative predictive value of elemental concentrations of U is 155 ppm with 11.30 error percentage, and for Th is 124 ppm with 8 error

percentage. The results described above have raised the prospect of further improvement of the technique developed so far its extension to analysis of other samples contain radioactive elements.

Suggestion

Further studies will still be needed to develop a laser ablation technique using more variation sample and sample standard needed to more precise qualitative results.

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