

Laser Induced Break down Spectroscopy (LIBS): A tool for identification of fuel pin failure by monitoring He gas

U.K.Maity, J. Namitha, P. Manoravi* and M. Joseph#

Materials Chemistry & Metal Fuel Cycle Group, Indira Gandhi Centre for Atomic Research, Kalpakkam – 603102, India.

#Raja Ramanna Fellow, IGCAR

(*Corresponding Author, Email: pmravi@igcar.gov.in)

1. Abstract / Introduction

The thermal heat produced during nuclear fission is transferred through the fuel-clad gap to the clad and the coolant is continuously extracting this heat. The gap between fuel and clad, plenum is filled with some conducting gases, called bonding gas. Helium is commonly used for its inertness, high thermal conductivity, neutron transparency and it does not form radioactive isotopes under reactor condition [1]. In a nuclear reactor, many safety mechanisms are incorporated for a safe operation of the plant and safe operating procedures are being practiced. In a fast reactor, one of the regular safety practice is the analysis of core cover gas for the detection of radioactive fission gases, such as Kr and Xe. This is being carried out towards the detection of failed fuel pin (FFP) at the inception stage itself. Conventionally, the detection of FFP is done by the delayed neutron detection (DND) method [2]. In this method, the delayed neutrons emitted from fission products are detected and this method is applicable only if there is a considerable breach in the fuel pin, which can allow release of fission products (delayed neutron emitter) into the coolant (liquid sodium) stream. However, in the event of very small leak in the fuel pin, in addition to gaseous fission products (isotopes of Kr and Xe), He (bonding gas) will come out. Other solid FPs will be retained inside the fuel pin. In such scenario, DND method cannot be employed. Hence, an effective detection of Kr or Xe or He in the Ar cover gas will indicate the failure of fuel pin. In this paper, online detection of He in the Ar cover gas is attempted using Laser Induced Break down Spectroscopy (LIBS). LIBS is a novel technique for elemental analysis based on laser generated plasma, irrespective of physical state of the sample. i.e. gas, liquid, solid, gel, etc. [3]. The laser pulse will ablate the sample and produce plasma plume, which expands resulting the de-excitation of atoms giving unique spectral signature, which are analyzed by a spectrometer. As indicated above, this present work is focused to determine the calibration curve for identification and quantifying the helium in argon atmosphere using LIBS.

2. Experimental technique

A modular 8 Channel fiber optic based LIBS (Model LIBS-8, M/s Applied Photonics, UK) covering wavelength range from 180 nm to 1030 nm is used for present studies [Figure 1]. Nd:YAG laser (Quantel Big Sky CFR 400) having 1064 nm wavelength emission, 10 ns pulse width, pulse energy of 390 mJ, and a focal spot size of app. 0.5 mm dia are employed in the LIBS. Two separate mass flow controllers are used for delivering the helium and Argon to the LIBS chamber. The flow rate of helium is varied between 175 sccm to 375 sccm with total flow of 800 sccm, corresponding to concentration variation from 21.8% to 46.8% of helium, respectively.

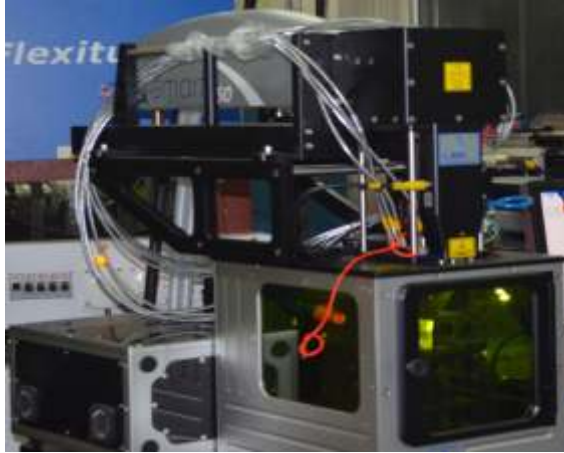


Figure 1: LIBS in Mini-cell

3. Results and Discussions

The LIBS spectrum contains emission lines of He, Ar (diluent) and common impurities (H₂, O₂, N₂, H₂O, etc.). The spectrum is analyzed to find the non-interfering emission lines of He. The selected lines are optimized for the laser energy of 390 mJ, signal accumulation for 400 laser shots and 20 Hz of repetition rate, under which the peak intensity is sufficient for analysis of lowest concentrations. He gas is diluted with UHP Ar such that He concentration varies between 21.8% to 46.8%. The non-overlapping emission peak of helium at 587.4 nm is used for the analysis [Figure 2]. The intensity of emission line for He is plotted as a function of He concentration [Figure 3]. The figure 3 indicates that the intensity of the emission line increases with the increase in concentration of He and the plot fits well linearly with correlation coefficient (R²) value of 0.992. The following equation is used to determine the limit of detection (LOD).

$$\text{LOD} = 3\sigma/S$$

Where ' σ ' and 'S' are the standard deviation of blank and slope of the calibration curve in Figure 3, respectively. Under such condition, the minimum He concentration that can be detected is 3%.

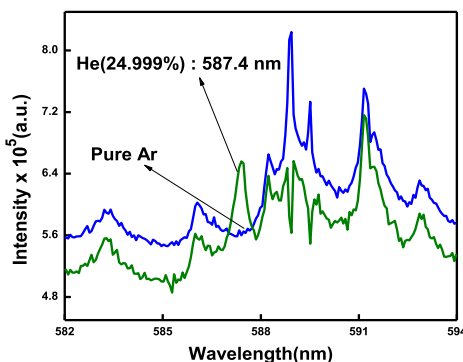


Figure 2. Non-overlapping emission lines for Helium

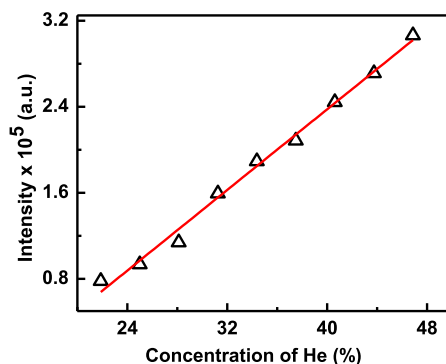


Figure 3. Calibration curve for Helium in Ar

Fuels for Indian fast reactors, namely Fast Breeder Test Reactor (FBTR), Prototype Fast Breeder Reactor (PFBR) are He bonded. In the event of a breach in the fuel pin, He will be released to cover gas. In such scenario, its expected concentration in cover gas will be about 0.4 ppm for both FBTR and PFBR. This concentration is much lower than LOD value of LIBS for He. He being an inert gas has higher ionization potential, 24.59 eV. Hence, LIBS with high power laser may possibly decrease the LOD value for He. Moreover, high resolution spectrometer will also contribute in this direction to lower the LOD value. In that case, LIBS will be potential technique for identification of failed fuel pin by monitoring He in cover gas. The main advantage of this method is that laser can be guide through portable fibre optics and the emission lines can also be collected and delivered to spectrometer through fiber optics, suitable for analysis at remote location.

Conclusion

LIBS is successfully employed for identification of He. Non-overlapping and highest intensity emission line for He is found at 587.4 nm. In the present work, He concentration is varied from 21.8% to 46.8 %. The calibration curve for He fits linearly with a good R² value of 0.992. The minimum detection limit for He in this method is about 3%. Further work is being explored to reduce the LOD value.

References

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