Development of Quick and Remote Analysis for Severe Accident Reactor-3
(2) Liquid-LIBS studies for the Fukushima Daiichi decommissioning

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Abstract
In the context of the Fukushima Dai-ichi Nuclear Power Plant (F1-NPP) decommissioning process, laser-induced breakdown spectroscopy (LIBS) has many advantages. The purpose of the present work is to demonstrate the on-line monitoring capability of the LIBS coupled with the ultra-thin liquid jet sampling method. The study focuses on zirconium and iron in aqueous solution, considering that these are major elements in the F1-NPP fuel debris.

Keywords: LIBS, solution, zirconium, iron, Fukushima Dai-chi decommissioning, ultra-thin liquid jet

1. Introduction

The laser-induced breakdown spectroscopy (LIBS) is used for the real-time, in situ and remote elemental analysis without any pretreatment. These advantages are very interesting for the post-accident environment analysis inside F1-NPS and for monitoring the contaminated aqueous solutions in decommissioning process. But it is difficult to use LIBS with liquid samples because splashes and ripples that form on the liquid prevent efficient detection of plasma emission in the ablation. However, using liquid jets can cost-effectively counter balance this issue. In this context, the present study focuses on the zirconium determination in aqueous solutions, as a major element of the debris material. Preliminary results obtained on iron - another major element of the fuel debris - will be shown also.

2. Experimental setup

The liquid recirculation system consists of a reservoir, a pump, and a slit-type nozzle equipped with XYZ motion and a rotation stage. The liquid jet of the sample solution is formed by a trapezoidal-shaped grooved nozzle tip composed of stainless steel with a nozzle exit of 0.6 mm × 0.3 mm. The pulsed Nd:YAG laser (fundamental) is focused on the thickest region of the liquid-sheet jet.

3. Results, discussion and conclusion

The LIBS experimental parameters were optimized for zirconium analysis in aqueous solution. For quantitative studies, the intermediate laser pulse energy (E_L) of 75 mJ/pulse provides a good signal to background ratio. Because of the atomic and ionic emission lifetimes differences, the optimum gate delay (t_d) were changed depending on the lines: t_d = 10 μs for the ZrII lines and t_d = 15 μs for the ZrI lines. The limit of detection could be decreased down to 4 mg L^{-1} in the case of the ionic lines.

Also, the preliminary results obtained for iron solutions shows that zirconium and iron should be analyzed simultaneously with moderate interferences.

References

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