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ARTICLE

Fundamental research on the integrated measurement method using laser and ultrasound for fuel debris investigation

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This study hopes to combine LIBS technology and ultrasound technology to quickly obtain the distribution, shape, and surface elemental composition of nuclear fuel debris in PCVs. In this paper, the working principle and the construction of the remote LIBS system are described. The operating principles of ultrasound systems for shape and solid-liquid surface distinction measurements are introduced. Then, the remote LIBS and ultrasound systems are integrated to develop an elemental mapping system. The validation experiment of the elemental mapping system is carried out on the simulated nuclear fuel debris. The elemental mapping system successfully obtained a 3D elemental map containing information on the simulated nuclear fuel debris samples' position, shape, and surface elemental composition. Finally, the shape measurement result from the ultrasound system was optimized using the elemental measurement result from the remote LIBS system. Through the validation experiment, it can be seen that the use of ultrasound measurements helps to obtain information about the shape and position and predict the solid and liquid surfaces of the simulated fuel debris simultaneously. This dramatically improves the efficiency of remote LIBS measurements. The application of the remote LIBS system can effectively measure elemental composition on the surface of simulated nuclear fuel debris. Meanwhile, the microchip LIBS system can optimize the shape measurement results of the ultrasound system. Therefore, integrating the two systems allows for more efficient and accurate measurement of nuclear fuel debris' shape, position, and surface elemental composition.

Keywords: ultrasound distance measurement; LIBS; solid-liquid surface distribution

1. Introduction

Over the years, the Japanese government has been promoting the decommissioning of the Fukushima Daiichi Nuclear Power Plant (FDNPP). An essential problem during decommissioning is the large number of radioactive nuclear fuel debris in the primary containment vessel (PCV). To decommission the FDNPP, it is necessary to remove nuclear fuel debris from the PCV. The shape, distribution, and element composition information of nuclear fuel debris is essential for accident analysis and the development of fuel debris removal plans. Currently, the shape and distribution of nuclear fuel debris in PCV are mainly investigated remotely by robots with optical cameras [1]. The element composition of nuclear fuel debris in PCV is primarily obtained by sampling off-situ element measurement. The robot arm collects and transports small nuclear fuel debris samples to the analysis center for element analysis. However, off-situ element measurement takes a long time, and it is impractical to analyze the whole PCV with this method.

Therefore, laser-induced breakdown spectroscopy (LIBS) and ultrasound technology were selected for this research. LIBS can perform in-situ element analysis and realize remote measurement using optical fiber [2,3]. The remote LIBS system is also suitable for installation on the robot arm because of its small size. However, this technology must focus the laser on the object's surface. Otherwise, the measurement accuracy will be reduced. Therefore, LIBS technology needs to determine the distance between the probe and the object. In addition, using robot arms to remove exposed nuclear fuel debris during the decommissioning process is also planned. Therefore, it is necessary to develop a method to simultaneously measure the distribution of nuclear fuel debris and coolant water levels. Ultrasound measurement is very suitable for measuring distance and shape. At the same time, ultrasound can determine the distribution of exposed nuclear fuel debris by distinguishing solid and liquid surfaces.

Therefore, this study hopes to combine LIBS and

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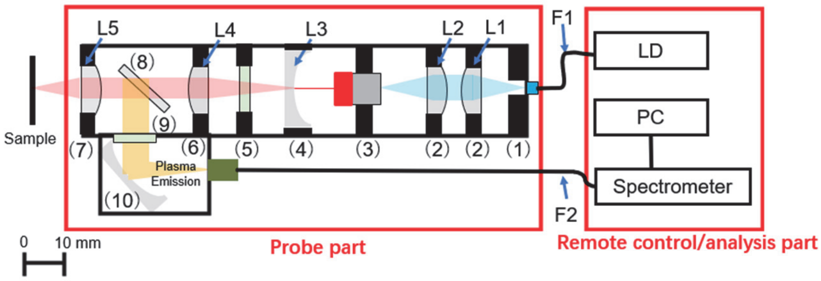

Figure 1. The diagram of the remote LIBS system.

ultrasound technology to quickly obtain the elemental composition and distribution on the surface of nuclear fuel debris in PCV. In this paper, an integrated system is built, and integrated experiments are carried out. The position, shape, and surface element composition of simulated nuclear fuel debris were successfully measured through experiments. The experimental results also show the integration of LIBS and ultrasound can make the ultrasound measurement results more accurate and improve the boundary detection between solid and liquid surfaces. Realizing an in-site elemental mapping system for fuel debris Investigation.

2. Construction of the integration experiment

2.1. Construction of remote LIBS system

2.1.1. Principle of remote LIBS system

LIBS is the laser-induced breakdown spectrum. Its working principle is to use the pulsed laser to generate the laser-induced plasma to excite the material in the sample to generate light. Then, the spectrometer is used to obtain the spectrum of light. By comparing the spectrum obtained with the spectrum in the LIBS database, the element composition of the sample can be identified.

In this research, the microchip LIBS was selected. This is because the high peak power laser required by LIBS may damage the optical fiber [4]. The optical fiber only transmits the low-peak power laser in the microchip LIBS system. At the end of the optical fiber, the microchip laser crystal converts the low-peak power laser to the high-peak power laser.

2.1.2. Setup of remote LIBS system

The diagram of the remote LIBS system is shown in **Figure 1**. LIBS system mainly includes two parts. The first part is the probe part -- the cage system. The second part is the remote control/analysis part, including a laser diode (LD), a spectrometer, and a PC. The microchip laser crystal is placed in the cage system. All the models used in the cage system are shown in **Table 1**. The main specifications of the laser diode are shown in **Table 2**. The main specifications of the spectrometer are shown in **Table 3**.

The low peak power laser is emitted from the LD and reaches the cage system via the optical fiber. In the cage system, the laser with low peak power is replaced by a microchip laser crystal with a laser with high peak power

Table 1. Models used in cage system.

Number (L: lens, Number: holder, filter, etc.)	Model (Thorlabs)
(1)	CP02/M, SM1SMA
L1; (2)	C230TMD-B; CP1M09/M
L2; (2)	C330TMD-B; CP1M09/M
(3)	SM1Z, SM1AD5
L3; (4)	LC2969-C; CPN06
(5)	NF808-34, DCP1A
L4; (6)	LA1560-C; CP14/M
L5; (7)	LA1540-YAG; CP14/M
(8)	DMLP950, CP360R/M
(9)	SM1PD2A
(10)	RC08SMA-F01, SM1A6, CP30
F1	SFH600M-SF22
F2	M15L02

Table 2. The main Specifications of the laser diode.

Specifications	Value
Maximum output voltage	2V
Maximum output current	160 A
Pulse duration	20.0 ~ 500.0 **µs**
Frequency	1~100 Hz
Output laser wavelength	808 nm

Table 3. The main Specifications of the spectrometer.

Specifications	Value
No. of elements	2048 pixels
Sensitivity	75 photons per count at 400nm 41 photons per count at 600nm
Pixel size	**14µm × 200µm**
Signal-to-noise ratio	250:1
Spectral resolution	0.035 nm

and focused on the sample. Then, the light emitted by the excited sample is received by the cage system and transmitted to the spectrometer for analysis via optical fiber.

2.2. Construction of ultrasound solid-liquid level measurement

2.2.1. Principle of ultrasound solid-liquid level measurement

Ultrasound solid-liquid surface measurement is divided into ultrasound shape measurement and solid surface and liquid surface distinction. The principle of ultrasound shape measurement is based on time of flight (TOF) [5,6]. The ultrasound probe sends an ultrasound wave to the object's surface, and the ultrasound wave will be reflected from the object's surface. Then, the ultrasound probe will receive the echo from the object's surface. The distance between the ultrasound probe and the object's surface can be calculated by measuring the ultrasound transmission time. The distance between the sensor and the estimated object equals half of the ultrasound propagation distance. The following formula expresses ultrasound distance measurement:

$$d = \frac{ct}{2} \tag{1}$$

Where d is the distance, c is the speed of sound, and t is the propagation time.

The principle of ultrasound solid-liquid surface distinction is that the fluctuation of the liquid surface will affect the intensity of the ultrasound echo. Before the measurement, the equipment drips water onto the surface to create water waves. When the ultrasound probe emits the ultrasound wave to the water surface, the ultrasound echo from the water surface will be disturbed by the water wave. The ultrasound wave's echo will be scattered, concentrated, or refracted in another direction. Therefore, the maximum amplitude of the echo changes significantly. On the contrary, the ultrasound echo from the solid surface will not change sharply with the water wave, so the maximum amplitude of the ultrasound echo is relatively similar. Therefore, the solid surface and the liquid surface can be distinguished by collecting five echoes at one spot and comparing the variance of the maximum amplitude of the echo from the liquid with the variance of the maximum amplitude of the echo from the solid surface.

2.2.2. Setup of ultrasound solid-liquid level measurement

The ultrasound solid-liquid level measurement system has been established based on the above principle. The diagram is shown in **Figure 2**. The system includes an air-coupled ultrasound probe for emitting and receiving the ultrasound. Data is transmitted to the PC through the pulse receiver and AD converter. The 3D stage is used to carry ultrasound probes and can move in three dimensions at 1mm intervals. The stage controller controls the 3D stage. The vessel contains water and samples. The water pump drops water into the vessel to generate water waves. The PC summarizes the ultrasound measurement data and the position information from the stage controller.

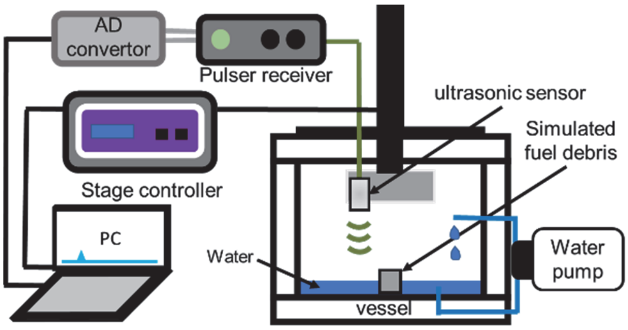

Figure 2. The diagram of the ultrasound solid-liquid level measurement system.

2.3. Construction of integrated experiment

2.3.1. Setup of the integrated experiment

The diagram of the integrated experiment is shown in **Figure 3**. The ultrasound probe from the ultrasound system and the cage system from the LIBS system are integrated into an integrated measuring unit. In the measuring unit, the ultrasound probe is 25mm higher than the LIBS cage system on the z-axis, 50mm larger on the x-axis, and the same on the y-axis. The integrated measuring unit is fixed on the 3D stage. The diagram and photo of the simulated sample are shown in **Figure 4**. The simulated sample is a cylinder with a diameter of 30 mm and a height of 31 mm. The white part in the middle of the sample is the compacted

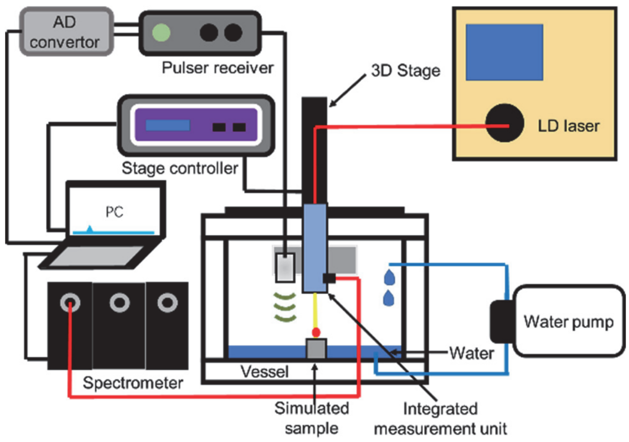

Figure 3. The diagram and photo of the simulated sample.

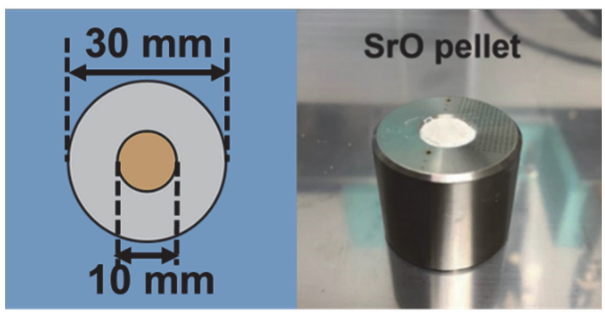

Figure 4. The diagram and photo of the simulated sample.

Table 4. The main parameters of the integration experiment.

Integration experiment setting	parameters
LIBS integration time (ms)	2000
LIBS scans to Average	1
LIBS boxcar width	0
LIBS electric dark	On
Ultrasound probe name	HAR0.4K 14×20N
Sound velocity (m/s)	340
Ultrasound probe sampling rate (S/s)	10M
Ultrasound probe center frequency (Hz)	400k
Ultrasound pulse repetition frequency (Hz)	100
Number of ultrasound repetitions	5
3D stage X-axis length (mm)	40
3D stage Y-axis length (mm)	40
3D stage pitch (mm)	1
Distance from the ultrasound sensor to the water (mm)	80

SrO powder with a diameter of 10 mm. The rest is stainless steel (SUS). The main parameters of the integration experiment are shown in **Table 4**.

2.3.2. Experiment steps of the integrated experiment

- First, add 5mm deep water into the container, place the sample in the container, and set the 3D platform at the starting point.
- Secondly, start the water pump to generate water waves. In this study, ultrasound sensors measured in two dimensions (X and Y axes) at 1 mm intervals and recorded five echo waveforms at each location.
- Thirdly, the solid surface can be inferred from the ultrasound measurement data, and the position and height of the solid surface can be determined.
- Fourth, move the 3D platform to the previously speculated solid surface area to start LIBS measurement. LIBS measures element composition information of two-dimensional (X and Y axes) at 2mm intervals.
- Finally, draw and analyze all the results with computer programs.

3. Results of the integration experiment

3.1. Results of ultrasound measurement

The result of the ultrasound shape measurement is shown in **Figure 5(a)**. The ultrasound shape measurement successfully reconstructed the shape of the simulated sample. However, the measured result is larger than the actual size of the sample. This is because ultrasound will disperse during transmission. So, the ultrasound will still hit the object's edge after the probe just left the top of the object. In this experiment, the spatial resolution of ultrasound shape measurement is 6 mm, and the temporal resolution is 10 ms. It can be reduced by replacing a smaller probe or placing it closer to the object.

The result of ultrasound solid-liquid surface distinction is shown in **Figure 5(b)**. In the figure, the pink dots are predicted to be solid surfaces, and the blue dots are predicted to be liquid surfaces. As shown in **Figure 5(c)**, the measuring area is divided into many squares with a side length of 5mm. A square with more than 60% solid spots is predicted to be a solid surface area and marked green. 60% is selected as the threshold because the ultrasound shape measurement's spatial resolution is 6 mm, and the square side length is 5 mm. So, if the squares include the sample, the measurement error can always ensure that the square will be judged as a solid area. So that there will be no omission in the later LIBS measurement. The experimental results successfully distinguished the solid and liquid surfaces. Some spots are misjudged as solid in the area that should be the liquid surface. This is because the viscosity of the water or the water wave is too small. It can be reduced by increasing the water wave. There are also some spots misjudged as liquid in the solid area. This is because the sample and probe are shaken when the 3D stage moves.

3.2. Results of LIBS measurement and elemental mapping

The result of LIBS element analysis is shown in **Figure 6**. LIBS system only conducts element analysis in the area judged as solid surface. This saved 26.6% of the time for LIBS element analysis in this research. The experimental result shows that LIBS element analysis successfully measured the surface elemental composition of the simulated sample.

By integrating LIBS measurement results with ultrasound

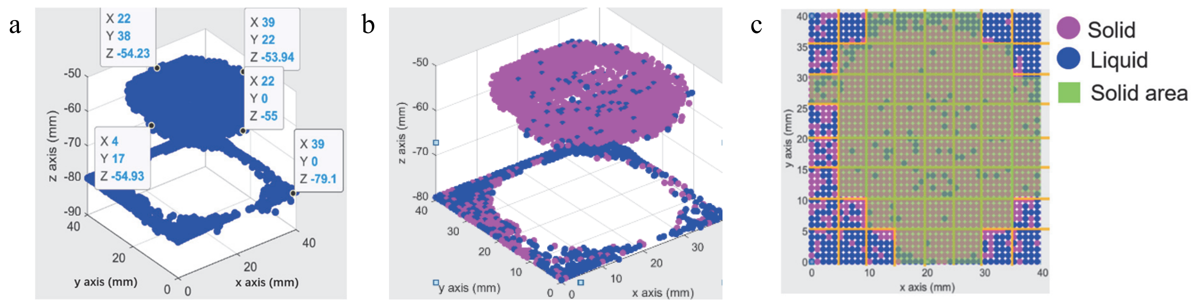

Figure 5. The ultrasound measurement result.
(a) The ultrasound shape measurement result, (b)The ultrasound solid-liquid surface distinction result, (c) Solid surface area predicted results.

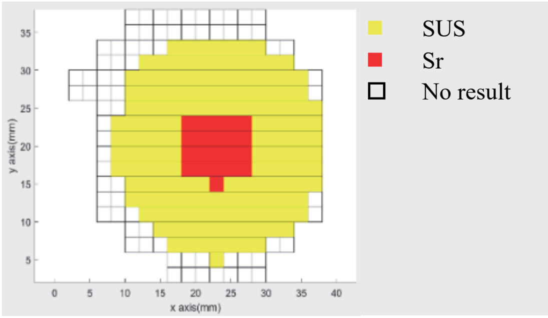

Figure 6. The results of LIBS element analysis.

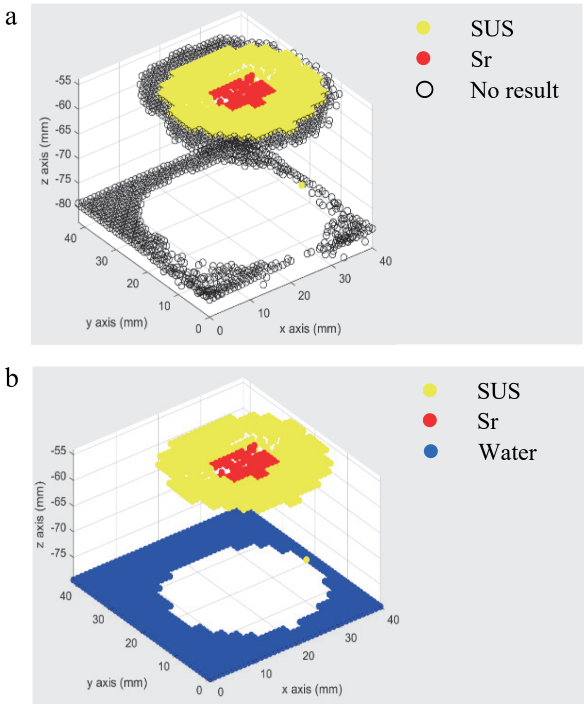

Figure 7. The original elemental map and optimized elemental map (a)The original elemental map, (b) The optimized result of the element map.

shape measurement results. We can draw the element map of the simulation sample, as shown in **Figure 7(a)**. And because the spatial resolution of LIBS measurement results is higher. It can significantly improve the accuracy of ultrasound shape measurement results. The optimized element map is shown in **Figure 7(b)**. The spatial resolution of the elemental map has been improved from 6 mm to 2 mm.

4. Discussion

Based on the above experiment results, integrating the remote LIBS systems and ultrasound system allows for more efficient and accurate obtaining of 3D elemental maps. Using ultrasound measurement helps to obtain information about the shape, position, and solid and liquid surfaces of simulated fuel debris simultaneously. This dramatically improves the efficiency of microchip LIBS measurements. The application of the remote LIBS system can effectively measure elemental composition on the surface of simulated nuclear fuel debris. Meanwhile, the LIBS results can optimize the shape measurement results of the ultrasound system.

However, it can be seen that the ultrasound system has significantly larger errors in the horizontal direction than that in the vertical direction. This is because ultrasound disperses as it travels. The beam width becomes larger. So, even if the sensor has left the object's top, some ultrasound pulses may still be hitting the object and producing echoes. What's more, the upper surface of the sample is closer to the sensor, so the echoes reflected from the upper surface have less distance attenuation. When the sensor receives a greater echo signal from the sample's upper surface than from the water surface, the ultrasound system might misjudge the distance, which is terrible for detecting the boundary of the simulated nuclear fuel debris. Therefore, optimizing the ultrasound shape measurement results with the LIBS measurement results is necessary.

Further research about the impact of other influencing factors on the system's accuracy is planned, such as the mist environment, water temperature, and surface cleanliness. Currently, the element mapping system cannot achieve underwater element and shape measurement, significantly limiting the system's ability to measure inside the reactor. Therefore, research on the improvement of underwater measurement is the focus of the future.

5. Conclusion

Integrating LIBS and ultrasound allows for the accurate and practical analysis of simulated fuel debris' surface elemental composition, shape, and position. Realized the in-site elemental mapping. The integration of LIBS and ultrasound can significantly improve the accuracy of ultrasound shape measurement. Further study on automation and using AI to identify element spectra will be needed to increase efficiency.

Nomenclature

d	distance from the ultrasound sensor to the object
c	speed of sound in air
t	propagation time of ultrasound pulses

References

[1] Tokyo Electric Power Company Holdings, On the investigation inside the containment vessel of Unit 1 reactor, *Decommissioning and Contaminated Water Response Team Meeting: 44th Secretariat Meeting* (2017), Tokyo, Japan.

[2] I. Wakaida, H. Oba, M. Miyabe, K. Akaoka, M. Oba, K. Tamura and M. Saeki, Laser-induced breakdown spectroscopy and related resonance spectroscopy for nuclear fuel cycle management and for decommissioning of "Fukushima Daiichi Nuclear Power Station, *Japanese Journal of Optics*, 48 (2019), pp. 13-20.

[3] Y. Deguchi, Development of Remote Elemental Analysis Using Laser Induced Breakdown Spectroscopy, *Bulletin of the Iron and Steel Institute of Japan*, 26

(2021), pp. 775-779.
[4] K. Tamura, H. Ohba, M. Saeki, et al., Radiation Dose Rate Effects on the Properties of a Laser-Induced Breakdown Spectroscopy System Developed using a Ceramics Micro-laser for Fiber-optic Remote Analysis, *Journal of Nuclear Science and Technology*, 58 (2021), pp. 405-415.
[5] R. Inoue, H. Tanabe, N. Shoji, H. Takahashi and H. Kikura, Object shape reconstruction technology using camera and ultrasound sensor, 49th Visualization Symposium (2021).
[6] T.V. Tran, H. Tanabe, H. Takahashi and H. Kikura, Visualization of Fuel Debris Shape and Measurement of Water Level using Parametric Ultrasound, The 13th Pacific Symposium on Flow Visualization and Image Processing (2022).