

# Collaborative R&D for Advanced Remote Analysis using Pulse Laser Ablation and Related Technologies

Akihiko Nishimura<sup>1,3,4</sup>, Yusuke Takenaka<sup>2</sup>, Tomonori Yamada<sup>1,3,4</sup>, Akinori Frusawa<sup>1</sup>, Takuya Shibata<sup>3,4</sup>, Shinichi Koyama<sup>4</sup>, Ikuo Wakaida<sup>3</sup>

<sup>1</sup> Applied Laser Tech. Inst., JAEA, Kizaki Tsuruga Fukui, 914-8585, Japan

<sup>2</sup> A-tech, Co. Ltd., Funaishikawa eki-nishi, Tokai-mura, Naka-gun, Ibaraki, 319-1116, Japan

<sup>3</sup> Collaborative Laboratories for Advanced Decommissioning and Science, JAEA

<sup>4</sup> Instrumentation and Laser Application Laboratory, JAEA

## Abstract

Collaborative activities promoting laser ablation and the related technologies are introduced. These technologies were transferred from Tsuruga site in Fukui to the new R&D site in Fukushima. Element Analysis by LIBS combined with a telescope and/or optical fiber delivery was demonstrated. And, deterioration Inspection by laser Doppler interferometer and micro debris sampling for mass spectroscopy and other advanced diagnosis were also arranged.

## 1. Introduction

We have been working over 7 years in Tsuruga site of Fukui to promote laser processing techniques for monitoring and maintenance of nuclear power plants (NPPs) [1], accompanying with the Human Resource Development [2]. The laser processing in JAEA can be originated to the project of laser isotope separation. While the ultrashort laser processing in JAEA could actually start in R&D for fiber Bragg grating, which is now under industrial deployment by Tsuruga Applied Laser Technology Institute [3].

Since the Great East Earthquake of Japan In 2011, decommissioning Fukushima Daiichi NPPs had been one of the most important issues for us [4]. Collaborative activities had started immediately after the accident in each establishment and all sectors of JAEA. Some special teams consisting of different research expertise were organized in the Sector of Fukushima Research and Development of JAEA. One of them challenged to develop a compact probing device in order to monitor the damaged reactor core of Fukushima Daiichi NPPs. The compact probing device was assembled, being designated as a prototype, applying pulse laser spectroscopy and telescope observation with radiation resistant optical fiber [5-6]. However, harsh environmental conditions of Fukushima Daiichi NPPs such as high dose radiation, high humidity and muddy water do not easily permit the use of robotic devices exploring in the concrete pedestal [7]. Thus, it will take a long time to succeed the remote analysis for the nuclear fuel debris.

For Fukushima reconstruction, Naraha Remote Technology Development Center (NRTDC) was established in 2015. The location is 30 km southern direction away from the Fukushima Daiichi NPPs [8]. Figure 1 shows a bird's eye view of NRTDC, where laser remote instrumentation of reactor decommissioning is opened for collaboration research. In addition, summer vacation internship program was arranged in Sep. 2016.



Fig.1 Bird's eye view of the Naraha Remote Technology Development Center

In this report, collaborative activities promoting laser ablation and the related technologies are introduced. These technologies were transferred from Tsuruga site in Fukui to the new R&D site in Fukushima.

## 2. Laser Ablation and Related Technologies

R&D for remote sensing technology for radioactive debris of Fukushima Daiichi NPPs is under promotion in NRTDC. We have prepared a laser laboratory in NRTDC where YAG lasers and a fiber laser can be used. Any laser experiment is controlled under the laser safety regulations. Here, laser ablation could be a core technology for handling radioactive materials. Figure 2 shows the relation of three major categories of R&D around laser ablation. The experimental setup is composed of a nanosecond YAG laser, focusing optics, a multi-channel spectrometer, a laser Doppler interferometer and a test piece of concrete sample. Nanosecond laser pulses are focused on the concrete surface. Then, laser induced ablation plasma is generated on a spot. A pair of YAG lasers is excited by a flashlamp at 20 Hz repetition. A high-power quasi-CW fiber laser can be also used as thermal ablation with millisecond laser pulses.

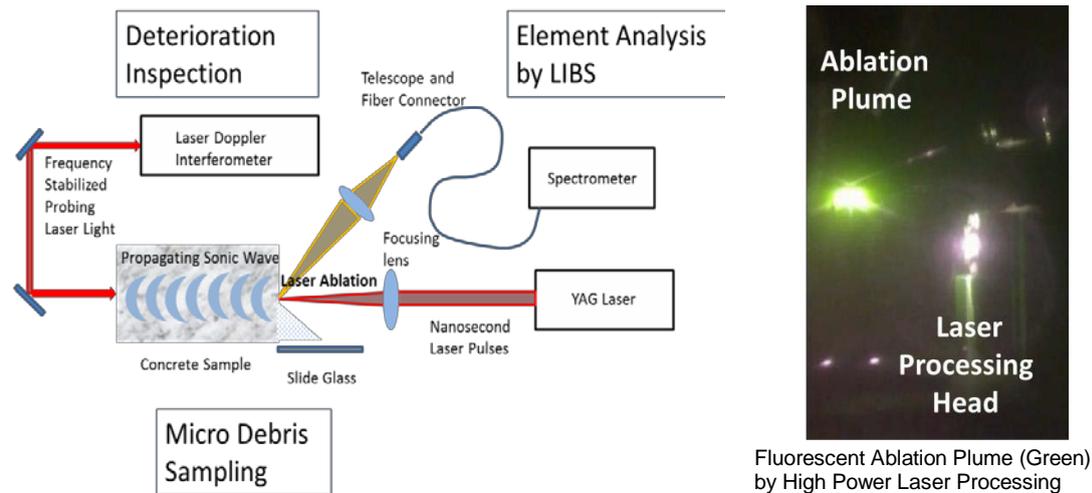


Fig.2 Three major categories of R&D linked with laser ablation (left); Laser ablation by a compact laser processing head (right)

## 3. Development of Laser Processing Heads

In Tsuruga site, we experienced the performance of laser welding by a high-power fiber laser. Figure 3 shows maintenance robotic devices which can access the limited space for welding, cladding and cleaning. A compact fiber laser was coupled with a laser processing head. An air-cooled 300 W CW fiber laser was used to repair cracks of welding beads. Motorized positioning stages lead it into the inside of a steel pipe. A composite-type optical fiber was used to deliver high power laser energy through a center core fiber. Endoscope image around the laser spot was transferred with counter direction. The composite-type optical fiber is unique to have coaxial bidirectional energy/image transportation. A feeding system for a filler wire was built on a circular mount plate.

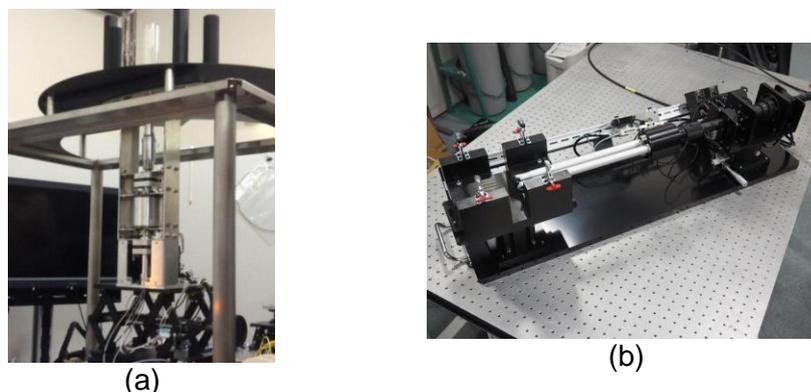


Fig.3 High power laser processing heads; (a) prototype laser maintenance device for a vertical heat exchanger, (b) advanced laser welding device for the butt section of steel pipes

Figure 3(a) shows a prototype laser maintenance device to access a vertical heat exchanger tube. The inner diameter is 1 inch so that the laser processing head was designed to have the size of a handy torch. Many improvement points were extracted by this device. The most troublesome point is how to avoid deterioration of mirrors and lens which are mounted in the laser processing head. Especially the high reflection mirror mounted at the head nozzle is in harsh condition so as to be exposed by radiation and fume from heating laser spot. Figure 3(b) shows the advanced laser device. Higher laser welding experiment was demonstrated to join a pair of butt pipes. A water cooled 6 kW CW fiber laser was used to check the performance of an optical fiber coupling unit. Laser welding was carried out at the butt section of a pair of steel pipes [9]. By EMAT sensing for the butt sections, laser energy of 1 kW is necessary to get penetration depth of the 3 mm pipe thickness. In order to reduce the deterioration of the high reflection mirror mounted on the head nozzle, gas flow rate in the laser torch is needed to keep 5 L/minute above.

#### 4. Remote Laser Processing Experiments

In Figure 2, the following three major categories of R&D linked with laser ablation are shown schematically. For the inspection for nuclear fuel debris of Fukushima Daiichi NPPs, the proof of principal of Element Analysis by LIBS using a radiation resistant optical fiber was finished successfully. To date, the robotic device which can carry a fiber coupled LIBS processing head into the reactor vessel is under planning. Deterioration Inspection is now under development. A laser Doppler interferometer was prepared to test deterioration inspection of heating effect on a concrete. We have specially fabricated concrete samples. The remote detection of surface vibration can measure the speed of propagating sonic wave through the concrete samples.

- 1) Element Analysis by LIBS combined with a telescope and/or optical fiber delivery
- 2) Deterioration Inspection by laser Doppler interferometer and numerical simulation
- 3) Micro Debris Sampling for mass spectroscopy and other advanced diagnosis

Micro Debris Sampling is still under preparation, the current status of which is reported in the next chapter. Laser ablation on the concrete samples is carried out. Fine particles of decomposed concrete are easy to be generated but are difficult to be gathered. There could be two methods for future development. One method is collecting relatively large particles. These particles are emitted from the laser spot in a parabolic trajectory and fall down around the laser spot. The other is micron-sized ultrafine particles floating in the air for a long time. Ultrafine particles easily condense due to humidity and static electricity. If it is assumed that the ultrafine particles are radioactive, it is important to control the airflow around the laser spot for safe handling of the ultrafine particles.

#### 5. Example of LIBS of a concrete sample

Q switched YAG laser pulses are focused on the sample surface by a focusing lens of FL=300 mm. Vaporized plume is rising so as to face the incident YAG laser pulses and becomes plasma and emits dazzling light. A sharp impact sound is produced when the plasma shrinks. The emission spectrum of a concrete sample is shown in Figure 4. A spectrometer, Ocean Optics HR 4000, is used. A telescope condenses light emission with a single lens of FL=50 mm and lead to the spectrometer with delivery optical fiber. The Q switch delay of the pulse laser was changed from 210  $\mu$ sec to 190  $\mu$ sec. The pulse energy is 8 mJ to 43 mJ. The strong peak at 1063 nm is the scattering of the pulsed laser light. The plume emits broadband light and shines white due to wide emission in the range of 250 nm to around 650 nm. Here, calcium ion lines of 393.366 or 396.847 nm are clearly observed in near UV region of Figure 4(a).

Figure 4(b) shows the enlargement of 700 - 780 nm. This range is less overlapped with spectral peaks of various elements and is effective for nuclear material measurement in nuclear reactors. However, since the peak intensity is low, sufficient integration time is required. Further, in the wavelength range of 700 to 900 nm, devitrification in the optical fiber

can be prevented even with integrated gamma ray irradiation of  $10^4$  Gy or more in case of high concentration OH group-doped quartz fiber. Therefore, it is important to confirm the emission spectrum of each element in advance in this wavelength range. Figure 4(b) shows three possible peaks of calcium atom of 714.82, 720.22 and 732.62 nm marked with blue arrows. The laser pulse repeats 2 Hz and the accumulated shot count is 16. These marked peaks are also obtained in LIBS experiments in water by optical fiber transmission. Since this experiment was carried out in air, it contains a peak derived from oxygen peaks of 777.19~777.54 nm.

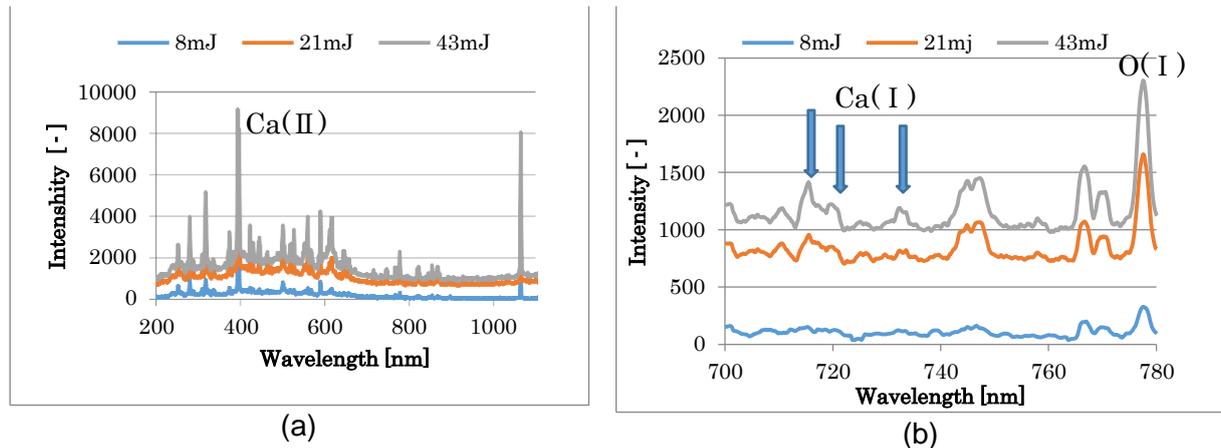


Figure.4 LIBS of a concrete sample; (a) wavelength range from 200 to 1100 nm, (b) wavelength range from 700 to 780 nm.

## 6. Current Status for Micro Debris Sampling

Laser drilling was on a concrete sample was demonstrated from the view point of structural health integrity for railway tunnels. An air cooled 1.5 kW Quasi-CW fiber was used in the experiment. It was investigated either in the downward or the upward direction. In case of concrete, melting expulsion was induced by recoil pressure evaporation and greatly enhanced by expansion via gaseous bubbles and breakup. The performance of laser induced melting exclusion was significantly enhanced in the upward direction by the assistance of gravity.

The performance by nanosecond YAG laser irradiation shows completely different aspect in comparison with that by Quasi-CW fiber laser irradiation. A thin layer of the concrete could act as a plasma confinement shell in laser peening process underwater. Higher recoil pressure of ablation plasma, several hundred MPa, is generated within a few nanoseconds. Flashing fluorescence and ejecting decomposed fine particles are accompanied with the ablation plasma. Thus, no melting glassy product is found around a laser spot.

We are starting a preliminary experiment as follows; A slide glass is set on an optical table just below the laser spot of concrete sample. Micro particles in the ablation plume rain down on a slide glass for microscope observation. The slide glass is coated with UV curable resin to make the micro particles confined strongly. Fine particles of decomposed concrete were captured on a slide glass and observed by a phase contrast microscopy.

The laser pulse energy was 8 mJ and the irradiation time was 10 seconds and 40 seconds. To increase recovery, laser pulse energy was increased to 43.4 mJ. Figure 5(a) shows a low magnification phase contrast microscope image with pulse energy of 43.4 mJ. Ultraviolet curing resin is applied to the left side of this image, and it is not done on the right side of the image. On the left side of the image, concrete fine particles collected in the resin are clear. There is a region with white contrast so as to surround the concrete micro-particles embedded in the resin. Since the polymer of the ultraviolet curing resin is oriented so as to surround the particle surface, it seems that the contrast is emphasized by the phase difference of the transmitted light with the randomly oriented region. Figure 5(b) is an enlarged image of the yellow border line in Figure 5(a). Fine particles with a diameter of several  $\mu\text{m}$  can

be confirmed. Ultrafine silica particles as the main component of concrete have been solidified by rapid cooling.

As shown in Figure 5(b), around 20  $\mu\text{m}$  diameter particles on a slide glass could be useful for LIBS because of the size far larger than the diffraction limit of a focusing lens. On the other hand, around 300 nm diameter ultrafine particles are rather applicable for efficient ionization of induction coupling plasma.

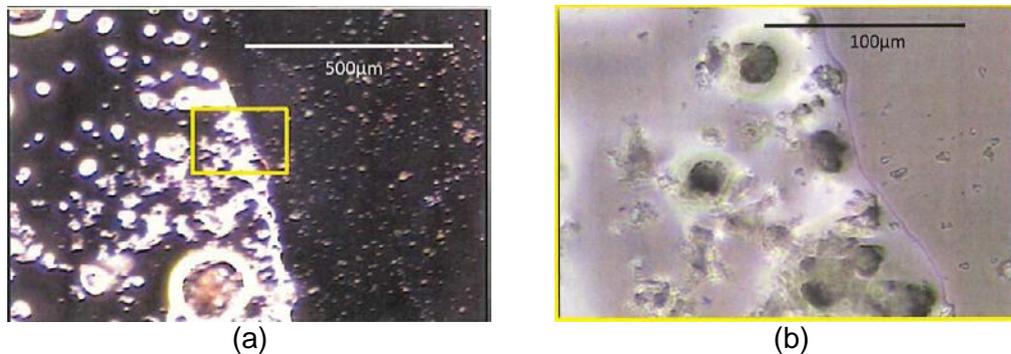


Fig.5 Fine particles of decomposed concrete sample by laser ablation; (a) Phase contrast image of UV curing resin, (b) Ultrafine particles of silica in the magnification image

## 7. Conclusions and Future Works

Three major categories of R&D linked with laser ablation, are important for remote processing and analysis for reactor decommissioning; Element Analysis, Deterioration Inspection and Micro Debris Sampling. Laser processing heads with proper functions packed in the compact body was successfully developed in Tsuruga site. Joining a radiation resistant optical fiber scope and a high resolution spectrometer to the laser processing heads, we are preparing a table-top telescopic LIBS system for demonstrations.

In Apr. 2017, a new laboratory was opened in Tomioka site of Fukushima. This will be the command tower that can handle collaborative R&D [10]. A few years later, in a hot laboratory of Okuma site of Fukushima, nuclear fuel debris and other contaminated objects will be handled and analyzed remotely. We are sure that pulse laser ablation and related technologies can contribute to not only reducing radiation exposure but also advancing analytical methods.

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