Development of a laser-driven ultrasonic technology for characterizations of heated and aged concrete samples

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1. Introduction

We experienced the Great East Japan Earthquake followed by the severe accident at the Fukushima Daiichi Nuclear Power Station, in which pieces of the molten nuclear fuel dropped down to the lower part of the Primary Containment Vessel (PCV) [1]. During this period, the concrete structures inside the PCV were exposed to a high-temperature environment [1,2]. Recovery of resolidified molten fuel mixed with the surrounding structures is called fuel debris; the development of remote inspection technology for degraded concrete structures is one of the key issues for safe and reliable decommissioning [2]. Such a remote inspection technology is useful not only for decommissioning but also for a wide range of aged infrastructures if the inspection technology is simple, reliable, and cost-effective.

Laser technologies have recently become increasingly practical; one of the attractive areas includes laser-based ultrasonic waves and their detection. Laser-produced plasma drives ultrasonic waves on the surface of a solid body [3,4], which passes through it, bringing information of macroscopic or mechanical properties of it. In this case, the laser intensity on the surface was more than 10\textsuperscript{7} W/cm\textsuperscript{2}, called the plasma regime [4]. However, a laser-based interferometric technology is used to detect ultrasonic waves driven by a laser illumination composed of the longitudinal and shear vibration modes where the former one is faster than the latter one [5]. The technology applies to the non-contact nondestructive testing of high temperature or molten metals [6–8] and various concrete structures [9,10], which we could not apply easily to the contact diagnostic systems.

In this paper, we propose measuring the velocity of ultrasonic waves and their spectra propagated through the concrete samples, which were functions of mass density – the elastic modulus of a concrete sample to develop inspection technology for judging its soundness.

To support the purpose, we showed the experimental characterization of laser-driven ultrasonic generation and its application to the nondestructive testing of concrete samples that had been heated and degraded. Various concrete samples were exposed in the specified high-temperature conditions with specified durations as concrete models experienced in the severe accident at the Fukushima Daiichi Nuclear Power Station.

2. Methods and materials

2.1. The laser-driven ultrasonic generation and the detection system

Figure 1 presents a schematic view of this technology. Ultrasonic waves traverse through the sample and drive a surface vibration when the waves reach the other end of the sample. In this configuration, a laser probe can detect the longitudinal mode of surface vibration with a velocity $V$. The frequency of the He–Ne laser probe is shifted by the Doppler shift $\Delta f$ as follows:

$$\Delta f = \frac{2|V|}{\lambda},$$

(1)
where λ represents the laser wavelength. The Doppler-shifted component is detected at the heterodyne detector using the Mach–Zehnder type heterodyne interferometer [11]. Finally, we obtained the time-dependent velocity and frequency of the longitudinal mode of surface vibration. Then, we achieved the time-dependent displacement with integration over a specific time duration.

One of the useful applications of this technology is the characterization of mechanical properties that depend upon the velocity of longitudinal waves measured by the traversal time of an object. The maximum velocity of the longitudinal wave c is represented as:

\[
c = \sqrt{\frac{E(1 - \mu)}{\rho(1 + \mu)(1 - 2\mu)}}
\]  

where E, μ, and ρ are Young’s modulus, Poisson’s ratio, and mass density of the sample, respectively. As shown in Equation (2), the velocity of the longitudinal wave is derived from Young’s modulus, Poisson’s ratio, and the Lamb’s constant [5]. Distinguished fastest signal contains macroscopic or mechanical properties of target material.

### 2.2. Experimental setup

Figure 2 presents the experimental setup. The ultrasonic wave was driven by a pulsed YAG laser, which can deliver energy up to 0.8 J in 6 ns pulse duration at 1.064 μm wavelength with a repetition rate of 10 Hz (Quantel Q-smart 850). The ultrasonic wave was detected by a few mW continuous wave (CW) high precision He–Ne laser beam at a wavelength of 633 nm, coupled with the Mach–Zehnder type interferometer and the heterodyne detection system (Polytech OFV-505 sensor head coupled with OFV-505 KA-LR controller) to measure the Doppler shifted component. Vibrations on the sample surface cause a shift on the laser frequency, and the shifted component was detected as a beat wave frequency at the heterodyne detection system. The amplitude and period of the beat wave represent the velocity and frequency of the surface vibration [11]. For detection, the laser beam was illuminated at the opposite side of the concrete sample, as shown schematically in Figure 1. We could measure the propagation time of the ultrasonic wave in the sample without contacting the sample.

Table 1 lists the composition of concrete samples prepared for this study. A mechanical strength associated with compressible stress to the concrete sample was 34.8 N/mm², which was similar to that used at the Fukushima Daiichi Nuclear Power Station [12]. The notation s/a* represents a volume ratio of fine aggregate/coarse aggregate. The part of coarse aggregate passed through the sieve with a nominal dimension of 15 mm, and the rest of the coarse aggregate was reassembled (mass ratio of 50 vs. 50). The notations of W, C, S, G*, AD, and AE stood for tap water, normal Portland cement, mountain sand, mountain gravel...
Table 1. Composition of the concrete samples. s/a* represented the volume ratio of fine aggregate/coarse aggregate.

| Water/cement | s/a* (%) | Water W (kg/m³) | Cement C (kg/m³) | Fine aggregate S (kg/m³) | Coarse aggregate G (kg/m³) | Chemical admixture AD (Cr%) | Chemical admixture AE (Cr%) |
|--------------|----------|-----------------|------------------|--------------------------|--------------------------|-----------------------------|-----------------------------|
| 55           | 45       | 157             | 285              | 838                      | 1035                     | 1.0                         | 0.0015                      |

Table 2. Heating, cooling rates, and holding time duration for each heating temperature condition. The temperature was measured at the center of each sample.

| Heating temperature (°C) | Heating rate (°C/min) | Cooling rate (°C/min) | Holding time (hours) |
|--------------------------|-----------------------|-----------------------|----------------------|
| 105                      | 0.48                  | 0.12                  | 504 (21 days)        |
| 200                      | 0.48                  | 0.28                  | 336 (14 days)        |
| 400                      | 0.64                  | 0.71                  | 24                   |

Figure 3. The mass density of concrete samples is shown as a function of the heated temperatures. The time history of each temperature exposed to the samples is listed in Table 2. The variation of each piece caused the error. Plot points denote the mean value obtained from two or three experiments. They were estimated based on Student’s t-distribution with a confidence level of 95%. Note that the errors of the solid circles without the error bars were within the circles.

(maximum size = 25 mm), air-entraining water reducing agent (standard 1st kind)/master poly zaglo15S, and air-entraining agent (1st kind)/MasterAir202, respectively. Concrete samples were heated and cooled for specified heating and cooling rates, and heating durations at the specified temperatures. These parameters are listed in Table 2. Note that the samples were heated slowly enough to almost keep the equilibrium temperature in the samples. The volume of the samples was measured using the electronic Vernier caliper to measure the lengths with an accuracy of ±0.04 mm (Niigata Seiki Co. Ltd. DT-300). The mass of the samples was also measured using an electronic scale with an instrumental error of ±0.04 g (SHIMAZU UW6200H). Figure 3 presents the measured mass density as a function of the heated temperature.

The physical and chemical processes of the heated concrete samples [13] are summarized as follows: volumes get expanded by heating the concrete samples, resulting in a decrease in mass densities and chemical changes; dehydration of calcium silicate hydrate and gypsum decomposition (CaSO₄·2H₂O), including water loss, occur in the first region of temperature between 100°C and 300°C; and, in the second region of the temperature between 300°C and 900°C, dehydration of calcium hydroxide occurs as:

$$\text{Ca(OH)}_2 + 1340 \text{ kJ} \rightarrow \text{CaO} + \text{H}_2\text{O(gas)}.$$  

The decomposition of calcium carbonate occurs as:

$$\text{CaCO}_3 + 1637 \text{ kJ} \rightarrow \text{CaO} + \text{CO}_2\text{(gas)}.$$  

Figure 4. A temporal shape of the ultrasonic wave is shown. Notations A and B represent the wavefront of vibration and the traversal time, including the intrinsic delay time of the instrument. The dotted line represents the trigger signal from the photodetector, shown in Figure 2. The velocity in the vertical axis was converted from the signal amplitude in volt, which is proportional to the velocity.
During this period, the gases (H₂O, CO₂) are released from a concrete sample [13]. These physical and chemical mechanisms are caused because of the variation of the mass density, as shown in Figure 3.

3. Experimental result

3.1. Temperature dependence of velocity of the ultrasonic waves

Figure 4 presents a typical ultrasonic wave. The abscissa represents the time axis, and the ordinate represents the velocity of the vibration given by the Doppler-shifted component, as shown in Figure 1 and Equation (1). We could identify the wavefront of vibration indicated as A in Figure 4 at the sample surface corresponding to the arrival time of longitudinal ultrasonic wave (p-wave). The traversal time through a sample and the internal delay were also indicated as B, as shown in Figure 4. Both measured wavefront of the vibration and the distance between the source of the ultrasonic waves and the detection point provide the velocity of ultrasonic waves as mentioned in the previous section.

To measure the velocity of the ultrasonic wave passing through a concrete sample, we used a retroreflective sheet to enhance the signal-to-noise ratio and make an accurate and stable measurement. Signals obtained with and without the retroreflecting sheet are shown in Figure 5(a,b), respectively. The sheet may discourage the real applications, although an order of magnitude higher ratio can be obtained. For practical use of this technique, we tried to use the higher power lasers, for example, a few W CW laser instead of few mW lasers; a signal level is expected to increase ~10³ times. In this case, we did not use any retroreflective sheet [14].

After the preparation, we successfully measured the traversal velocity through the concrete samples. The velocity as a function of the heated temperature listed in Table 2 is shown in Figure 6. The velocity decreased at 105°C centigrade due to the water loss; it dropped rather significantly at 400°C due to the change of chemical compositions (described in the previous section), causing a change in the mechanical property. Figure 7 shows a list of the ultrasonic waveforms and corresponding spectra.

When the samples are heated at 105–400°C, the number of peaks in the spectra increases compared with that of unheated samples. We expect ultrasonic waves to reflect inside the samples due to heating effects. The number of cracks in the samples exposed to higher temperatures is increased, and the ultrasonic waves are more frequently scattered. Although spectral information may provide useful information, it does not give us clear quantitative information on the material properties in these experimental conditions. However, the velocity of the ultrasonic waves provides us clear information on the material properties.
Figure 7. Waveforms and their Fourier transformed spectra of the ultrasonic waves as a parameter of the heated temperatures. (a)–(d) correspond to the room temperature (20°C), 105°C, 200°C, and 400°C, respectively.
Figure 8. The displacement of the vibration as a function of time. Each signal was made with numerical integration of the velocity waveform over time, as originally shown in Figure 7(a-d).
We also estimated the displacement of the vibration at the detection surface of the sample as follows:

\[ X(t) = \int_0^t f(t) \, dt, \quad (3) \]

where \( t \), \( X(t) \), and \( f(t) \) represent time, the time-dependent displacement of vibration, and the time-dependent velocity, respectively. We have already obtained all these. For example, the calculated amplitude of the vibration as a function of time is shown in Figure 8. Figure 8(a,c,e,g)

are a typical displacement of the vibration. Notably, low-level direct current (DC) components were found in the velocity signals, which were as small as 3.1–4.6% of the peak amplitude of the velocity signals. The DC components in the velocity signals caused a linear increase in the displacement signals with vibration components. According to the observed velocity signals, the samples were found to approach the driving laser, while this physical phenomenon is not possible. Therefore, the DC component was thought to be caused by the electronic system. We subtracted the DC components in the velocity signals, as shown in Figure 8(b,d,f,h). The magnitude of the vibrating components in the displacement signals was typically 0.02–0.04 µm under the present experimental conditions.

### 3.2 Dependence of the velocity on the laser-focused condition on the samples

We set a sample at a translational stage to change the sample position systematically and to measure the velocity as a function of the irradiated surface position of the YAG laser where the laser-focused position was fixed. Figure 9 shows the velocity as a function of the YAG laser irradiation position. The position of S represents the standard irradiation and is placed at the laser focus position on the surface of the samples. B represents the place where an air breakdown happens; in that position, the laser beam is focused on the air, and the measured velocity drops down.

Figures (10,11) present the snapshots of air breakdown indicated in Figure 9. Figure 10 shows a full view of the breakdown recorded using a video camera with a frame rate of 30 frames/s. Figure 11 also shows the breakdown with much higher temporal resolution recorded by a high-speed camera (Photon FASTCAM Mini AX50) with a frame rate of 5000 frames/s. The emission from the breakdown region was growing in time; however, it was shortened along the laser propagation axis after maximizing the region.
Figure 12. Ultrasonic waveforms and their spectra correspond to the data in Figure 9. In (d), the laser-driven air breakdown happened.
3.3 Dependence of YAG laser pulse energy on the velocity of the ultrasonic waves

We investigated the traversal time of the ultrasonic waves through an unheated sample as a function of YAG laser pulse energy, as shown in Figure 13. The velocity was almost insensitive to the laser pulse energy.

The signal amplitude at the wavefront indicated in Figure 4 as a function of YAG laser pulse energy is shown in Figure 14. The signal amplitude tends to be almost proportional to the laser pulse energy. The laser pulse energy of more than a few hundred mJ was considered desirable for the measurement due to suitable signal levels in the present experimental conditions.

4. Discussions

The velocity of the longitudinal wave was represented in Equation (2), as shown in the previous section. We should take into account the mass density correction shown in Figure 3: the Lamé’s constants, which are a function of Young’s modulus, Poisson’s ratio, bulk modulus, and modulus of rigidity for calculating the velocity of the longitudinal wave. When the temperature increases, the mass density becomes smaller, but it is less than 6.3% compared with that of a sample kept at the room temperature. Figure 15 shows the measured compressible strength obtained with JIS A 1108 and the static elastic modulus obtained with JIS A 1149 by the measured compressive strengths for the concrete samples. When the heated temperature increases, the decrease of the elastic modulus is thought to be caused by the creation of number of cracks and the chemical decompositions. According to Eq. (2) and Figure 15, the decrease of the elastic modulus leads to the decrease of the velocity of the ultrasonic waves, as shown in Figure 6.

From this point of view, the surfaces of the samples were observed with a digital microscope (Hirox Co. Ltd. KH-8700), as shown in Figure 16(a–d). When the samples were heated at the temperature of 105°C, the relatively small and larger number of cracks were observed on the surface as shown in Figure 16(b) compared with a surface of an unheated sample as shown in Figure 16(a). The widths of the crack were <9.8 µm. The small cracks were presumed that hydrated regions shrank with dehydration. In addition, the result of X-Ray Diffraction test showed clear peaks of Ca(OH)\(_2\) at the temperatures of 105°C, 200°C, and 400°C, as shown in Figure 17. It represents that almost no chemical decomposition of cement paste occurred below 400°C. While the peaks of Ca(OH)\(_2\) were significantly reduced when the heated temperature was 600°C [15] and it was expected that the chemical decomposition started to occur over 400°C.

When the concrete samples were heated at 200°C, the widths of cracks are <16.0 µm, as shown in Figure 16(c). When the concrete samples were heated...
at 400°C, the decomposition occurred and the aggregate expanded. Simultaneously, the cement hydrate regions shrink and its inhomogeneity causes self-straining stress, which is thought to create more cracks and reduced strength [16]. The widths of cracks were <48.9 µm, as shown in Figure 16(d). The cracks are expected to distribute almost homogeneously in the samples because each component of the concrete is distributed homogeneously in the samples and the number and size of the cracks are increased by heating. These results show that the vibrational frequency is affected by the increase in the number of cracks in the concrete samples, as shown in Figure 7. When the samples were heated at 400°C as shown in Figure 7(d), spectral components of vibrational frequency of >100 kHz decreased significantly compared with that of unheated samples as shown in Figure 7(a). Moreover, lower frequency spectral components did not decrease significantly in the samples exposed at the higher temperature atmospheres.
5. Conclusions

We proposed to measure the velocity as a function of mass density and the elastic modulus of the concrete sample through its samples to develop inspection technology for assessing its soundness. To support this proposal, we have performed an experimental characterization of laser-driven ultrasonic generation and its transport through the concrete samples that have been heated and degraded. For this purpose, the various concrete samples were exposed in the specified high-temperature conditions with defined temporal profiles as concrete models experienced in a severe accident at the Fukushima Daiichi Nuclear Power Station. We could successfully obtain the experimental results by measuring the velocities of an ultrasonic wave passing through the concrete samples. The velocities and spectra of the ultrasonic waves are strongly dependent on the exposed temperature, i.e., at a high-temperature condition of 400°C, the velocity was 3700 m/s, and at room temperature, the velocity was 5000 m/s. The measurement of velocities of an ultrasonic wave with the present technology provides almost the same result as with the contact ultrasound technology [15]. The present technology shows the applicability of nondestructive testing of degraded concrete structures. It is noted that in the actual Fukushima Daiichi Nuclear Power Station, significant parts of the concrete structures have been exposed to water, where the strength of the concrete has been expected to recover from the initial condition, which was caused by the severe accident [17,18] Further studies on variety of experimental conditions are needed to apply this technology to the decommissioning of Fukushima Daiichi Nuclear Power Station. The technology is also expected to contribute to the maintenance of a wide range of concrete structures.

Supplementary file

We also tested the samples heated at 600°C, 700°C, and 800°C with the same experimental configuration. As described previously, when the temperature increased above 600°C, the mechanical property changed significantly. We expect that it is very difficult to detect transmitted ultrasonic waves, because the waves are significantly attenuated because of severe thermal deterioration of concrete and the creation of cracks, as shown in Figure A1. However, we detected the vibration signals, as shown in Figure B1. The spectra are concentrated in the low-frequency region of < 50–60 kHz compared with those shown in Figure 7. The origin of the signals could not interpret as ultrasonic waves. Measured signals were stable and reproducible. Signals came from the surface vibration, probably caused by the vibration of the sample body driven by the laser irradiation. Figure C1 shows the compressible strength obtained with JIS A 1108 and the static elastic modulus obtained with JIS A 1149, as presented information for readers.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

References

[1] The Fukushima Daiichi accident, description and context of the accident. Austria: International Atomic Energy Agency. 2015 [cited 22 OCT 2019]. Available from: https://www-pub.iaea.org/MTCD/Publications/PDF/AdditionalVolumes/P1710/Pub1710-TV1-Web.pdf.
[2] Technical strategic plan 2018 for decommissioning of the Fukushima Daiichi nuclear power station of Tokyo electric power company holdings, Inc. Japan: Nuclear Damage Compensation and Decommissioning Facilitation Corporation. 2018 [cited 22 OCT 2019]. Available from: http://www.dd.ndf.go.jp/en/strategic-plan/book/20181109_SP2018eOV.pdf.

[3] Hutchins DA, Dewhurst RJ, Palmer SB, et al. Laser generation as a standard acoustic source in metals. Appl Phys Lett. 1981 May;38:677–679.

[4] Davies SJ, Edwards C, Taylor GS, et al. Laser-generated ultrasound: its properties, mechanisms and multifarious applications. J Phys D: Appl Phys. 1993 Mar;26:329–348.

[5] Krautkramer J, and Krautkramer H. Ultrasonic testing of materials. 2nd ed. New York: Springer-Verlag Berlin Heidelberg; 1977. Part A, 1. Waves 1.4 Formulas and Numerical Data; 17–21.

[6] Scruby CB. Some applications of laser ultrasonics. Ultrasonics. 1989 July;27:195–209.

[7] Burgess K, Prakapenka V, Hellebrand E, et al. Elastic characterization of platinum/rhodium alloy at high temperature by combined laser heating and laser ultrasonic techniques. Ultrasonics. 2014 Apr;54:963–966.

[8] Fuse N, Kaneshige K, Watanabe H. Development of thickness measurement system for hot steel with laser-ultrasonic wave technology. Mater Trans. 2014;55:1011–1016.

[9] Shimoda M, Oomura H, Misaki N, et al. Development of non-destructive inspection method for concrete material with laser-induced vibration. RTRI Rep. 2009 Dec;23:29–34. [in Japanese].

[10] Shimada Y, Kotyaev O. Remote sensing of concrete structures using laser sonic waves. Indus Appl Laser Remote Sensing. 2012;153–169.

[11] Johansmann M, Siegmund G, Pineda M. Targeting the limits of laser Doppler vibrometry. All Polytec. 2005:Page 1 of 12–Page 12 of 12.

[12] The report on the investigation into the current seismic safety and reinforcement of the reactors at Fukushima Daiichi Nuclear Power Station (No.1). Japan: The Tokyo Electric Power Company, Inc. 2011 [cited 28 APR 2020]. Available from: https://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110528e16.pdf.

[13] Hager I. Behavior of cement concrete at high temperature. Bull Pol Ac Tech. 2013;61:145–154.

[14] Kurahashi S, Shimada Y. Development of fast inspection techniques for internal defect in concrete structures with laser driven ultrasonic wave techniques. Annual Progress Report 2018–2019 of the Institute for Laser Technology. 2019 Jul; 32–36 [in Japanese].

[15] Luu VN, Murakami K, Do TMD, et al. Applicability of ultrasonic-wave based method for integrity assessment of concrete severely damaged by heat. E-J Adv Maint (Jpn Soc Maintenology). 2020;11:163–171.

[16] Abe T, Furumura F, Tomatsuri K, et al. Mechanical properties of high strength concrete at high temperatures. J Struct Constr Eng. AJJ. 1999 Jan;64:163–168. [in Japanese].

[17] Poon C, Azhar S, Anson M, et al. Strength and durability recovery of fire-damaged concrete after post-fire-curing. Cement Concr Res. 2001 Sept;31:1307–1318.

[18] Li Z, Ryuda Y, Sugihara D. Change and recovery of properties of fire-damaged concrete after re-curing; mass, length, dynamic elastic modulus and ultrasonic velocity. Cement Sci Concrete Technol. 2015;69:363–370. [in Japanese].
Figure A1. Cracks seen on the concrete surface. (a)–(c) correspond to the 600°C, 700°C, and 800°C, respectively.
Appendix B

Figure B1. Waveforms and their Fourier transformed spectra of the ultrasonic waves as a parameter of the heated temperatures. (a)–(c) correspond to 600°C, 700°C, and 800°C, respectively.
Appendix C

Figure C1. Compressive strength (rectangles) and static elastic modulus (circles) as a function of heated temperature. Plot points denote the mean value obtained from three experiments. Error bars are estimated based on Student’s t-distribution with a confidence level of 95%. Error bars with solid circles and solid rectangles without the bars are within the size of the symbols.