Study on acoustic detections of Nd:YAG laser induced breakdown at different wavelengths

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Abstract. Laser-induced breakdown detection (LIBD) is among many techniques utilizing laser beam to characterize nanoparticles in solution. It is based on the detection of plasma formation resulted from the interaction between particles and laser beam. In acoustic LIBD, the acoustic signal which accompanies the plasma formation is detected by a microphone. In this study, a 1064 nm pulsed laser was used as the laser source for LIBD system instead of the frequently used 532 nm. The presence of colloids in filtered and unfiltered water was investigated using the new system and the result was compared with those from conventional system. The S-curves from 1064 nm system showed similar trend with S-curves from 532 nm system. The presence of larger nanoparticles in unfiltered water resulted in lower breakdown threshold energy. However, the breakdown threshold energy of both filtered and unfiltered water was much higher in the 1064 nm LIBD system. The shifting was attributed to the high absorbance of water at 1064 nm. This research proved the possibility of using 1064 nm pulsed Nd:YAG laser as an alternative wavelength source for LIBD system, nevertheless various setup arrangement might be needed.

1. Introduction
In recent years, nanotechnology is getting a bigger role in science. Smaller nanoparticles are being synthesized. Along with this trend, more and more advance characterization techniques for nanoparticles are being developed. Laser-induced breakdown detection (LIBD) is one of them. Being able to detect and quantify small colloids in low concentration, LIBD is known to be more sensitive compared to other quantification techniques [1,2]. LIBD also needs no special treatment for the preparation step, i.e. a liquid sample can be measured as it is [3].

LIBD based its detection technique on the plasma generation resulted from interaction between a focused, pulsed laser beam and nanoparticle in solution [4-7]. Laser irradiation induces the so-called breakdown of the dielectric properties of a given medium [8-10]. Breakdown threshold, which is the power density needed to induce breakdown, is lowest for solid, higher for liquid, and highest for gases [8,11]. This difference is used to detect solid nanoparticles presences in solution. Breakdown threshold also differs from one particle’s size to another. Smaller nanoparticles needs higher power density for a breakdown to occur; therefore, it has a higher breakdown threshold [7]. The presence of more nanoparticles also results in higher breakdown probability, which is the statistical evaluation of breakdown occurrences [7].

Common setup for LIBD involves a 532 nm Nd:YAG laser as a pulsed beam source, a focusing system, a cuvette filled with liquid sample, a detector, and a data acquisition and processing system.
There are two types of breakdown detection, acoustic detection and optical detection [12]. In acoustic detection, a microphone is used to detect the acoustic signal generated along with plasma formation, whereas a charge-coupled device (CCD) camera is used in optical detection to capture the image of the plasma itself. Other types of pulsed laser source have been used in some LIBD experiments, such as a 500 nm excimer-dye laser and a Q-switched 532 nm diode-pumped solid state (DPSS) laser [13]. However, there is still not much information on whether LIBD technique can be performed at different wavelength. An Nd:YAG laser has a 1064 nm wavelength at its fundamental and there are various experiments carried out at this wavelength, e.g. laser-induced breakdown spectroscopy (LIBS) or synthesis of nanoparticles [14,15]. Combining 1064 nm LIBD with other experiments using 1064 pulsed laser will increase the research efficiency.

In this research, LIBD experiment was carried out by the proposed setup using 1064 nm pulsed laser and also the conventional setup using 532 nm pulsed laser. The breakdowns were detected acoustically by a microphone and the resulting breakdown probabilities were compared.

2. Experimental

The experimental setup for acoustic LIBD is shown in figure 1. Pulsed laser beam produced by a nanosecond Nd:YAG laser (Q-Smart, 10 Hz) was focused into the middle part of liquid sample contained in a cuvette. Once pulsed laser beam interacted with colloid, plasma was generated and followed by a breakdown. A microphone connected to a recording system recorded the accompanying acoustic signal. Recording was set to accommodate one measurement in an audio file. Each file was processed and analyzed using a homemade processing program [16]. It is also possible to record several measurements in one audio file since the homemade program provides the option to determine which part of an audio file that to be analyzed. However, it might slow down the data processing since it was heavier and calling such an audio file requires more time.

In this study, two different laser wavelengths were used: (i) the fundamental wavelength of 1064 nm, and (ii) the second harmonics 532 nm. The energy was set in intervals such that it covered from zero breakdown to maximum. Q-Smart is able to provide a maximum energy of 850 mJ at 1064 nm and 430 mJ at 532 nm. Triplicate measurements were carried out, each employed 1000 laser pulses. Water samples were prepared in two conditions: filtered and unfiltered with filtration performed at 0.22 μm. To prevent contamination, a glass cover was placed on the top of the quartz cuvette. The operational pulsed laser energy was measured considering the attenuation by glass cover.

3. Result and Discussion

Observation during LIBD experiment showed that the interaction between infrared (1064 nm) pulsed laser beam and nanoparticles resulted in the formation of plasma, which was accompanied by acoustic
signal emission. This was to be expected since 1064 nm pulsed laser has been frequently used in LIBS experiment to determine the elemental constituents of a sample based on its plasma emission [14]. The acoustic emission has been well detected and recorded by the data acquisition system. Evaluation by the homemade data processing system returned the number of breakdown events. Division by the number of laser shot resulted in the value of breakdown probability [12]. Figure 2(a) showed the S-curve plot of the resulting breakdown probability of filtered and unfiltered water against pulsed laser energy at 1064 nm wavelength.

![Figure 2](image)

**Figure 2.** The resulting S-curves of filtered and unfiltered water from the 1064 nm LIBD experiment (a) which can be divided into three regions (b).

The S-curves from 1064 nm LIBD experiment exhibited the trend of a typical acoustic LIBD S-curve [7]. Breakdown probability scales linearly with the effective focal volume, which means more ‘target’ for laser beam shot in a larger focal volume [12]. Focal volume is proportional to pulsed laser energy [12]. Therefore the breakdown probability increased as the pulsed laser energy was increased. Figure 2(b) displays the three regions of interest: threshold, slope, and saturation region. The threshold region marks the adversity in generating plasma due to low pulsed laser energy. At the end of this region lies the breakdown threshold energy, which is like a ‘bottleneck’ of breakdown probability. It represents the generation of ‘first electron’ in the multi photon ionization process where the value depended on the material and the detection process [12].

In the second region, the curve formed a linear slope, which corresponded to the linear range of focal volume as a function of pulsed laser energy [12]. Accordingly, this slope is concentration dependent. The slope of LIBD S-curve can be used to determine the particle concentration in a liquid sample. A higher number of particle has a higher slope and vice versa. The linearity ends near absolute (a probability of 1). At this level of photon flux density, an increase in pulsed laser energy did not result in a higher number of breakdown events and therefore ‘saturation’ occurred. In this acoustic LIBD, it was assumed that the maximum number of breakdown that might occur due to one laser pulse is 1 [12] and so saturation took place when nearly all of pulsed laser shot had produced breakdown. The acoustic LIBD did not recognize multiple breakdown events (two or more plasma generation by one pulse laser shot).

Figure 2 (a) also implies the ability of 1064 nm LIBD system to differentiate the average size of nanoparticles or colloids presented in the samples. Filtered water sample had a higher value of breakdown threshold energy which showed the smaller colloidal size compared to that of unfiltered water sample. Filtered water samples contained colloids whose average size was smaller than 0.22 μm
since the filtration process removed those larger than 0.22 μm. Accordingly, the unfiltered water sample also contained colloids larger than 0.22 μm which gave a rise in the average size value. Smaller colloids require a higher photon flux density to ignite breakdown because the total cross-sections for multi photon ionization is proportional to the particle volume (i.e. the number of valence electrons in the particle) [3,17], hence laser irradiation on filtered water sample ignited breakdown at higher energy and the S-curve of filtered water sample was located on the right side of the unfiltered water sample.

Despite its ability to perform LIBD measurement in the same manner with 532 nm system, 1064 nm LIBD system exhibited a quite different trait in term of breakdown threshold energy. Figure 3 shows the shifting in breakdown threshold energy as a result of changing the 532 nm LIBD system into the 1064 nm LIBD system. The experiment revealed that the 532 nm pulsed laser beam ignited breakdown at relatively low energy, i.e. below 5 mJ; whereas breakdown ignition by 1064 nm pulsed laser occurred at much higher energy, in the order of ten. This phenomenon is attributed to the different light absorbance in water for those two wavelengths. Previous studies on optical characteristics of water [17,18] reported that water has lowest attenuation, i.e. lowest absorbance, at around 500 nm. At 1064 nm, the water attenuation is almost two order of magnitude higher. In another word, a laser beam with 532 nm wavelength is better transmitted by the water than the 1064 nm laser. At 1064 nm, photon were attenuated by the water which resulted in significant energy decrease by the time it interacted with colloids. Additionally, energy per photon also contributes to the shifting of breakdown threshold energy. The energy carried by one photon at 1064 nm is much lower than that carried at 532 nm. Both water attenuation and energy per photon would affect the generation of the ‘first electron’ in multi photon ionization process. Therefore at 1064 nm, a much higher pulsed laser energy was set in order to compensate the lost.

There are various experiments that are often performed by 1064 nm Nd:YAG laser. This experiment result showed that an LIBD system might be set-up alongside those experiments and carried out alternately without having to change the laser source and redo the alignment. Furthermore, it may also be possible to integrate the LIBD system into those other experiments, such as laser-induced breakdown spectroscopy (LIBS) and nanoparticle synthesis. For example, an LIBD measurement may be performed during the synthesis of nanoparticle without having to change the laser source. Such an integrated system may be able to provide data on the nanoparticle formation and stability in a more efficient way.

![Figure 3. LIBD experiment using 1064 nm and 532 nm pulsed laser resulted in breakdown threshold energy shifting.](image-url)
4. Conclusion

It was possible to used 1064 nm pulsed laser for LIBD experiment. The laser was able to ignite plasma emission in the water sample. The number of breakdown occurrences increased as higher pulsed laser energy applied. The trend form an S-curve, a particular trait of LIBD result, was clearly observed. The 1064 nm LIBD system was also able to distinguish sample's particle size from each other based on the difference in breakdown threshold energy. Filtered water, which contained only small size colloids, had higher breakdown threshold energy than unfiltered water, which also contained bigger size particles. However, higher pulsed laser energy was needed in the 1064 nm LIBD experiment, compared to that in the 532 nm. Optical absorbance of water and energy per photon were mostly accounted for this phenomenon. Overall, the LIBD experiment using 1064 nm pulsed laser was a success. However cautions must be taken in such experiment, both for the system tools and users, due to the involvement of higher energy and invisible laser beam.

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