Breakdown Ignition of Nonsolvent Ionic Liquid with Double Pulse Laser

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1 Introduction

Recently, a low toxicity, high combustion quality, and high-density specific impulse have become important in the field of liquid propellants for spacecraft engines, and various studies related to them are actively underway. All of these studies aim to decrease the cost and to improve the operability of a rocket or satellite. With this background, we have studied the application of an energetic ionic liquid (EIL) based on ammonium dinitramide (ADN) as a liquid propellant [1]. This EIL consists of three solid components and is generated through eutectic melting. With the characteristics of these components, this EIL can attain a higher energy density. Table 1 summarizes a comparison of two EILs and hydrazine, a conventional liquid propellant.

Table 1: Propellant Properties

		Hydrazine	ADN-ELIPs(442)	ADN-ELIPs(631)
Density[g/cc] @20℃		1.0	1.5	1.5
Freezing Point [℃]		1.4	0	25
Specific Impulse [s]*		233	250	285
Adiabatic Flame Temperature [K]*		871	1986	2640
Toxicity	LD50 Oral [mg/kg]	60	-	-
	LD50 Transdermal [mg/kg]	91	-	-

*Calculation Condition : P_c = 0.7 [MPa], A_e/A_t = 50

An ionic liquid has several characteristics including a very low vaporability, which contributes to the high storability. From the viewpoint of combustion, this characteristic means that an ionic liquid has a low ignitability and incombustibility. This EIL includes ADN, a high energetic material. If this ELP ignites

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and burns, its adiabatic flame temperature during combustion will reach over 2000 K according to an NASA-CEA calculation [2]. This temperature easily destroys a conventional igniter system, e.g., a catalyst or spark plug, which are contact systems. Therefore, it is necessary to investigate a new ignition method for ELPs. In our study, we focused on laser breakdown ignition with a pulse laser, which transfers energy in a noncontact manner and has a high energy density. There are some of reports of laser breakdown ignition; however, nearly all of them are related to gas target breakdown [3]. When an EIL is sprayed by an injector, most of this spray is in the liquid phase because of the low vaporability of the EIL. For the ignition of EILs, it is important to know the behavior of an EIL droplet by pulse laser irradiation. Our group observed the droplet behavior with one laser pulse under several conditions with a high-speed camera and an ICCD camera and investigated whether a wider beam size atomized and gasified EIL droplets efficiently [4][7]. According to the pressure profile, this phenomenon was not a combustion but similar to decomposition. From these results, we consider the use of double pulse laser breakdown ignition. The first pulse may atomize and decompose the gas; then, the second pulse will break down the decomposed gas and ignite it. In this paper, we report the results of double pulse breakdown ignition of EIL droplets.

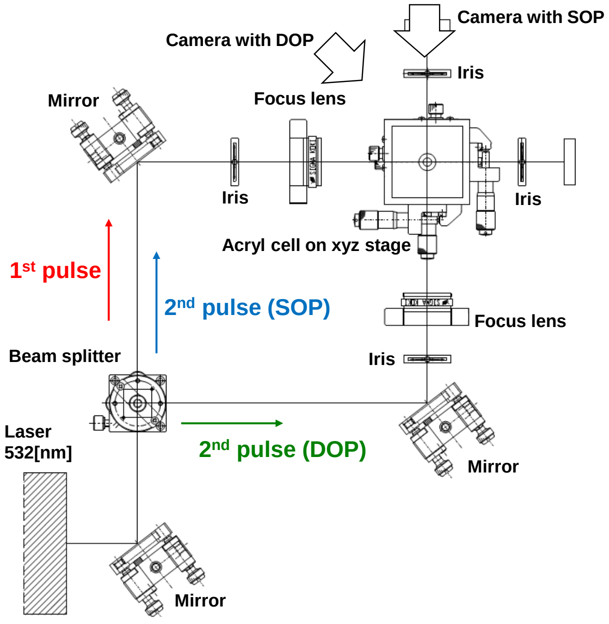

Figure 1. Diagram of experimant equipments

2 Experimental Procedure

This experiment utilizes a PIV-400 laser system (Sirah, pulse rate H=10 Hz, pulse width 8 ns, max laser power I_{max} = 620 mJ/pulse, beam diameter D_{beam} = 9 mm). The PIV-400 laser system can generate two independent pulses because it consists of two Nd:YAG rods with a controller. In this study, we choose a delay generator (DG535, Stanford Research Systems) as the external laser controller. From previous work, the first pulse is necessary for defocusing to increase the beam size to be greater than the EIL droplet diameter. In this study, we use a focus lens with focal length f = 100 (NADL-30-100PY2, Sigma Opt. Inc.) and set it at the f = 90 position for the first pulse, which is the defocus condition. Our group reported that beam density 3.8×10^{10} W/cm^2 is suitable for atomization and gas generation with one pulse [4]. We adjusted the first pulse power to 75 mJ, and the second pulse power was fixed at 40 mJ/pulse because it is the lowest stable power to induce the breakdown of air at 1 atm. One microliter of an AND-based ionic liquid droplet (ADN:monomethylamine nitrate (MMAN):urea = 40:40:20 wt%) was suspended on a glass rod (ϕ = 0.1 mm) in an acryl cell. If we use same optical path for first and second pulses, the breakdown point due to second pulse is not located at the droplet center. Therefore, it may better to control the focus conditions of the first and second pulses independently. We tested two pattern methods for double pulse breakdown ignition in N_2 at 1 atm and compared the results. One is that the first and second pulses pass through the same optical path (SOP), and the other is that they originate from different paths (DOP). Figure 1 shows a schematic of experimental equipment.

In the SOP mode, we use one optical path; thus, the breakdown point of the second pulse deviated from the expected position. Therefore, the injection timing of the second pulse must be optimized. The laser irradiation is controlled by a delay generator connected to the laser system. In the DOP mode, a beam splitter is selected to separate the laser beam. This method creates two beam paths for each pulse. It is essential to discuss the atomization and gasification effects of the double pulse inserted in only the first pulse shot. However, from previous reports, it seems that the defocused pulse effect is clearly larger than the focused pulse. We use a half mirror (HBCH-20-550, $I_{trans} = 45 \pm 5\%$, Sigma Opt.) as a beam splitter, which increases the pulse power to two times higher than that in the SOP mode.

In both experiments, we evaluated phenomena with a high-speed camera (Photron, AX-200, 100000 fps), 105 mm F4UV lens (Nikon), and band-pass filter (Asahi Opt., $\lambda < 500$ nm). This filter was selected to measure the combustion spectra of OH* ($I_{\max}$ at 306 nm) and CH* ($I_{\max}$ at 410 nm) [5]. In the SOP and DOP modes, the camera was located at positions 90° and 45° from the first pulse path, respectively. The pressure profile was measured in parallel with a pressure sensor (XT-140, Kulite Inc.) and an amplifier (AM32AZ, Unipulse).

3 Results and discussion

SOP mode

For the irradiation timing of the second pulse, it is important to know the behavior of the droplet due to only the first pulse shot. Figure 2 shows this behavior. At 0 ms, plasma luminescence occurs, and the droplet spreads in the opposite direction of the laser with time. Such behavior was reported by Klein [6], who discussed the droplet shape during laser breakdown. Comparing our results and those in Klein's report, our results show a high speed and wide spreading of droplet due to the defocus effect and higher laser energy. According these results, we carried out four experiments with delays between the first and second pulses of 5, 10, 100, and 1000 μs. As an example, we show the experiment with the delay of 100 μs in Figure 3. In these experiments, we could insert the second pulse by adjusting the timing for atomizing the droplet. A comparison of the flames before and after the second pulse shows that there are no major differences. Moreover, we did not observe chemiluminescence, as in the combustion spectra between 300 nm to 400 nm. The focal point of second pulse is 10 mm from the original position because the focus lens $f = 100$ is set at the $f = 90$ position. The breakdown point may be a somewhat far from the gas and droplet area. In addition, the second pulse propagates from the other side on which the droplet spreads. If a pulse is injected into the chamber, the atomized droplet and related gases cause laser scattering and prevent laser focusing. As a result, combustion was not observed in these experiments. From the pressure measurement, we did not observe a rapid increase such as combustion or ignition. Therefore, the second pulse for gas breakdown should be inserted from the other side of first pulse path.

DOP mode

In this case, it is necessary to set the timing of the second pulse and its position. The droplet mostly spreads on the opposite side from which the laser propagates (Figure 2). In this study, the second pulse is located 1–5 mm to the left of the droplet center on the first pulse path. First, we observed the droplet behavior in the DOP mode using only the first pulse to adjust the second pulse timing (Figure 4). The first pulse has a higher effect on atomization than the second pulse, which is focused. Then, we used a double pulse with a different optical path. The results for a second pulse having a 3 mm displacement and 100 μs delay are shown in Figure 5. Gas breakdown occurred at the adjusted positon at 0 and 100 μs. The breakdown at 100 μs may contribute to combustion of the EIL or derivative gases, but we did not observe a flame or chemiluminescence. After 100 μs, the behavior is not different from that during the insertion of only the first pulse. In other words, the probability that this method causes the EIL to burn is low. The pressure measurement did not indicate a rapid increase, as in combustion or ignition. Unlike the SOP mode, the second pulse in the DOP mode propagates in a different direction relative to the spread of the droplet; therefore, the effect of laser scattering is low. Therefore, there are other reasons for the lack of ignition and combustion. We reported that the gasification efficiency is less than 10% [7]. One reason may be that the concentration of the decomposed gas is below the limit of ignition. In this case, we have to develop effective methods for decomposing the ionic liquid.

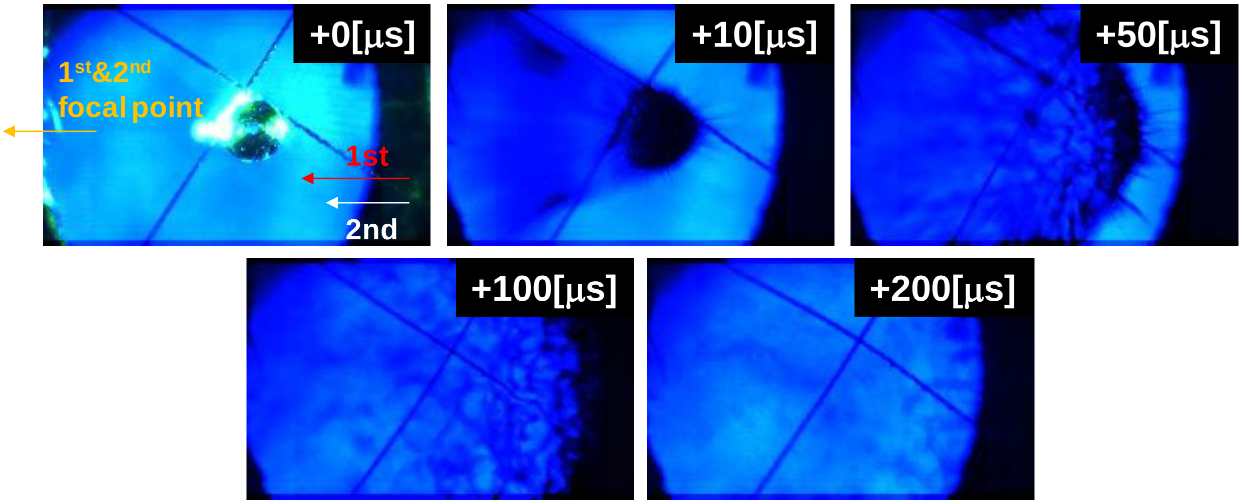

Figure 2. Droplet behavior for only the first pulse (75 mJ/pulse), 0 μs = laser irradiation time.

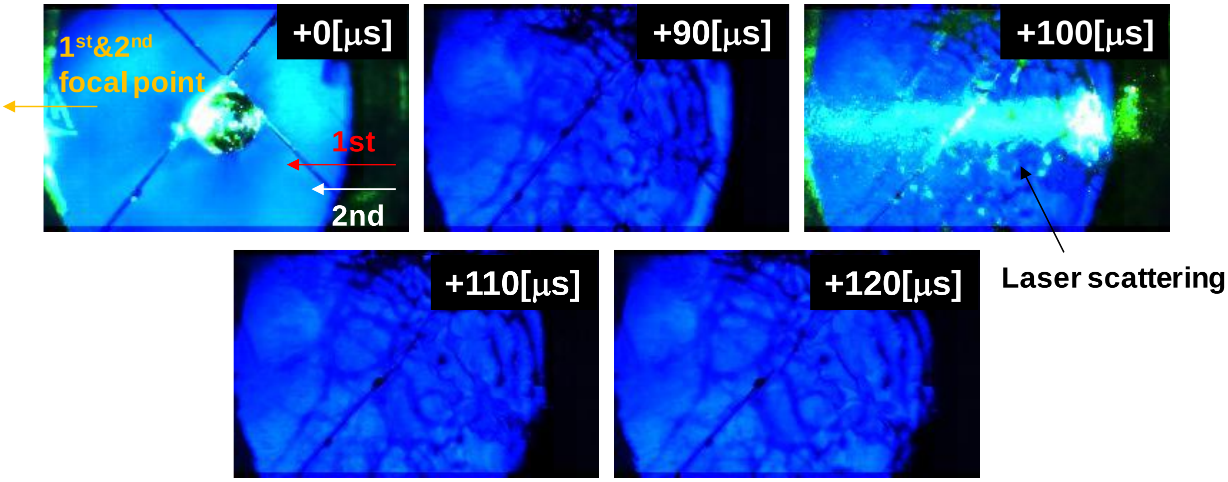

Figure 3. Droplet behavior for the double pulse (75 and 40 mJ/pulse) in the SOP mode with a 100 μs delay.

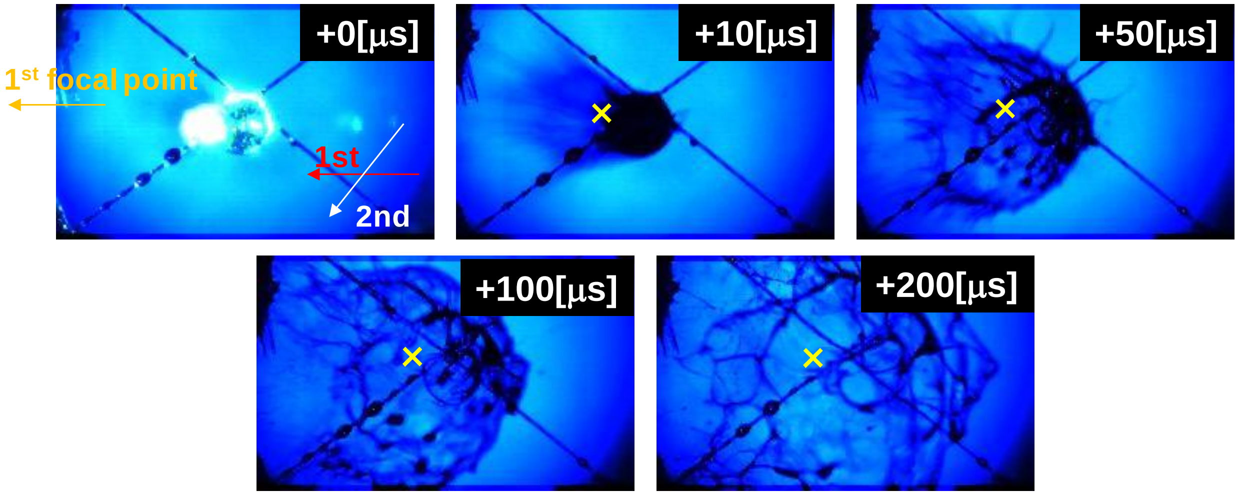

Figure 4. Droplet behavior for the double pulse when only the first pulse is separated by a splitter (150 mJ/pulse); the yellow cross is the focal point of the laser, 1 mm from the droplet center.

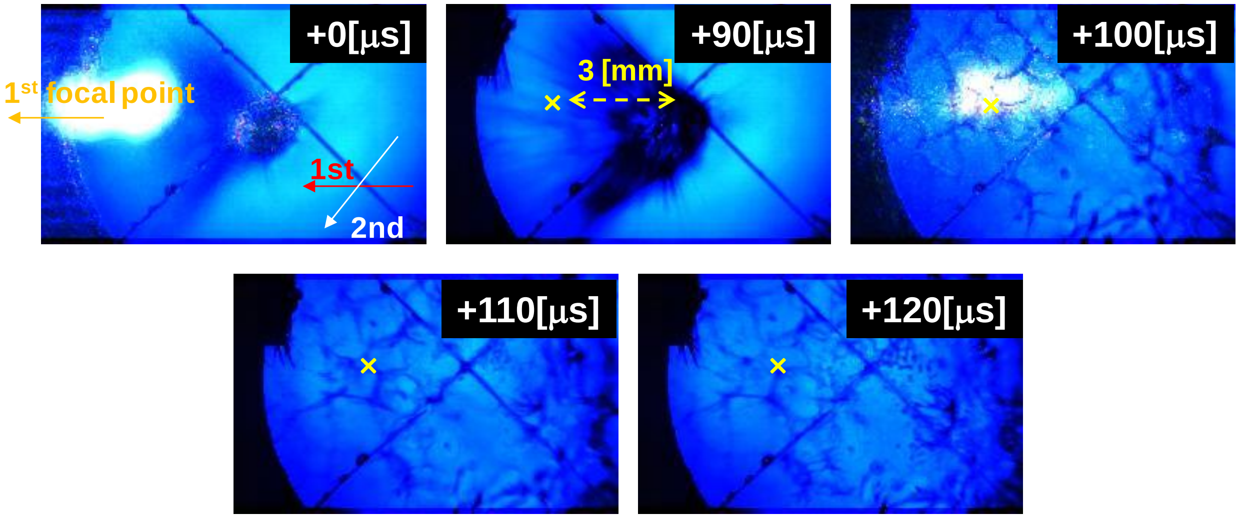

Figure 5. Droplet behavior for the double pulse (150 and 90 mJ/pulse) in the DOP mode; the yellow cross is the focal point of the laser.

4 Conclusion

We carried out two methods of double pulse breakdown ignition for the combustion of EILs based on ADN. In the SOP experiments, we could see the droplet behavior with irradiation with one pulse and setting the emission timing of the second pulse. The atomized droplet and decomposed gases scattered the second pulse and prevented the concentration of laser light. In the DOP mode, we optimized the laser irradiation timing and focal point of the second pulse. For both methods, we did not observe any behavior similar to combustion. It is necessary to develop a heating system for the gasification of an ionic liquid, which has an extremely low volatility, to ignite it.

5 Next Step

In this study, we used only 1 μL of an ionic liquid, and the generated gas volume is dependent on the droplet size. We should consider the ignition limit, pressure, temperature, concentration, and gas species. Further, it may be suitable to calculate the laminar burning velocity by CHEMKIN-PRO and compare it with the reported values. Moreover, it is important to increase the droplet volume and perform the double pulse breakdown experiments again.

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