



Optical ignition of nanoenergetic materials: The role of single-walled carbon nanotubes as potential optical igniters

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ABSTRACT

We demonstrate the effective optical ignition of nanoenergetic materials (nEMs) by adding single-walled carbon nanotubes (SWCNTs) as an optical ignition agent into an nEM matrix composed of Al/CuO nanoparticles (NPs). The measured burn rate and pressurization rate of SWCNT (optical igniter)-Al NP (fuel)-CuO NP (oxidizer) composites were found to attain maximum values of ~ 240 m/s and $\sim 5.9 \pm 0.8$ psi/ μ s, respectively, in the case of SWCNT (1 wt.%)–nEM composites. As the amount of SWCNTs added into the nEM matrix was further increased (i.e., SWCNT > 2 wt.%), both the burn rate and the pressurization rate of the as-prepared SWCNT–nEM composites decreased significantly. This was because the large amount of SWCNTs in the nEM matrix increased the heat dissipation through the SWCNT agglomerates, so that the temperature increase in the SWCNT–nEM composite was limited, thus hindering the oxidation and subsequent ignition of the nEMs. This suggests that the remote optical ignition and controlled-explosion reactivity of nEMs can be performed by incorporating SWCNTs as a potential optical ignition agent with nEMs.

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1. Introduction

Nanoenergetic materials (nEMs) composed of nano-sized fuel and oxidizer components can rapidly convert chemical energy into thermal energy when they are ignited. The ignition of nEMs can be performed using various means such as an electrical spark, flame, and hotwire igniters [1,2]. These means of ignition require direct contact between the igniter and the nEMs because the thermal ignition of nEMs is effected through the direct heat transfer from the external igniter to the nEMs. This approach also represents a limitation of the potential applications of nEMs in various thermal systems. Unlike thermal means of ignition, optical ignition of nEMs can provide many advantages, in that ignition can be achieved remotely, and the process is less sensitive to surrounding environmental factors, such as temperature, pressure, space, and time. A laser beam is one representative approach to the optical ignition of nEMs [3]. However, it generally requires high power consumption and complex mechanical/electrical auxiliary systems. Therefore, to easily ignite nEMs with a simple optical device, we have focused on the use of a flash of light instead of a laser beam.

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A flash of light has a wavelength of 400–1000 nm [4]. In our approach, the estimated energy density of the flash was approximately ~ 2 J/cm², based on the energy stored in the capacitor (1300 μ F, 350 V) of the flash circuit, and its average output reaches ~ 2000 W/cm² for a flash duration of 1 ms when there is direct contact between the flash and the nEM sample [5,6]. One might also need to employ an ignition promoter, which can easily be made to react with the irradiation of a relatively weak light source. As a potential optical ignition promoter in an nEM matrix, single-walled carbon nanotubes (SWCNTs) present a very attractive possibility. It has been reported that SWCNTs were unexpectedly ignited when they were exposed to a camera flash [7–10], which took place because heat was generated by the light absorption of SWCNTs and was subsequently transferred along the tubes to catalysts (e.g., Fe, Ni, etc.). The repeated irradiation of a flash of light on SWCNTs triggered a local exothermic reaction at the surface of the catalysts and finally ignited a bundle of SWCNTs.

In this work, we aim to investigate the effect of SWCNT addition in an nEM matrix on the optical ignition and explosion characteristics of nEMs. We specifically consider a thermite mixture of aluminum nanoparticles (Al NPs; fuel) and copper oxide nanoparticles (CuO NPs; oxidizer). We demonstrate that the characteristics of the optical ignition, as well as the burn rates and pressurization rates of nEMs in which SWCNTs have been incorporated can be significantly altered by controlling the amount of SWCNTs that is added into the nEM matrix.

2. Experimental

Commercially available Al NPs (NT base Inc.) that had an average primary particle size of approximately ~ 80 nm were used as a fuel. CuO NPs (Sigma Aldrich) that had an average primary particle size of ~ 50 nm were used as an oxidizer. SWCNTs (Hanwha Nanotech Inc.) grown on catalytic Fe nanoparticles were used as an optical ignition controller. We prepared SWCNT-nEM composites for a series of optical ignition tests, as shown in Fig. 1a. First, Al NPs (fuel) were mixed with CuO NPs (oxidizer) in an EtOH solution. Here, the mixing ratio of fuel and oxidizer was fixed at Al:CuO = 3:7 (wt.%). The SWCNTs were then added to the nEMs precursor solution (i.e., Al NP/CuO NP/EtOH) in mixing ratios of 1, 2, 5, and 10 wt.%, respectively. The Al NP/CuO NP/SWCNT solutions were dispersed by a sonication process (200 W, 40 kHz) for approximately 40 min and then dried in a convection oven at 80 °C for 30 min. Finally, the SWCNT-nEM composites were obtained. The resulting SWCNT-nEM composites were then characterized by various methods, including scanning electron microscope (SEM; S4700, Hitachi) operated at 15 kV and Cs-corrected scanning transmission electron microscope (STEM; JEM-2100, JEOL) operated at 200 kV. The SWCNTs were also analyzed by thermo gravimetric analysis (TGA; Pyris 1, Diamond, Perkin Elmer) at temperatures ranging from 30 °C to 800 °C at a heating rate of 10 °C/min in air. To examine the role of SWCNTs as a potential ignition controller, we also performed a series of flash ignition tests for SWCNT-nEM composites, as shown in Fig. 1b and c. In brief, we placed SWCNT-nEM composite powder on a glass slide under which a conventional camera flash was installed. We recorded the entire burning process of the SWCNT-nEM composite powder using a high-speed camera (FASTCAM SA3 120 K, Photron) at a frame rate of 30 kHz.

To investigate the effect of the SWCNTs on the explosion reactivity of the nEMs, the pressurization rates of the SWCNT-nEM composites were measured using a pressure cell tester (PCT) system [11]. In brief, an SWCNT-nEM composite powder sample fixed at 13 mg was placed in a closed cell that had a constant volume (~ 13 ml); it was then ignited by a heated tungsten wire that was placed on top of the powder, which was coupled with an external DC power supply operating at 2 A and 1.5 V for resistive heating. The pressure induced by the resulting explosion was measured in situ by a piezoelectric pressure sensor (Model No. 113A03,

PCB Piezotronics) attached to the pressure cell. The detected pressure signal was then amplified and transformed into a voltage signal by using a series of in-line charge amplifiers (Model No. 422E11, PCB Piezotronics) and a signal conditioner (Model No. 480C02, PCB Piezotronics). The resultant signal was recorded using a digital oscilloscope (TDS 2012B, Tektronix).

3. Results and discussion

Figure 2a and b, which present images of the SWCNTs employed in this approach, clearly show that the SWCNTs are bundled to some extent, and that they have an average diameter of ~ 1.5 nm. To examine the ratio of catalysts to CNTs in the SWCNTs, we performed TG analysis for the SWCNTs, as shown in Fig. 2c. It was observed that the initial mass of the SWCNTs decreased significantly as the temperature increased, due to the oxidation of the SWCNTs, and then remained at approximately 30% of the original sample mass at a temperature ranging 500 °C–800 °C. In order to examine the residual materials after TG analysis of SWCNTs, XRD and TEM analyses were performed as shown in Fig. 2d and e. X-ray diffraction patterns show very strong peaks corresponding to crystallized Fe_3O_4 (Fig. 2d). After heating SWCNTs at 800 °C in TG analysis, the SWCNTs were completely oxidized and the catalytic particles were grown by coalescence, ranging from 10 to 25 nm in diameter (Fig. 2e). This suggests that the SWCNTs employed in this study were composed of 23% Fe catalysts, 7% oxygen component, and 70% SWCNTs. The optical ignition of a bundle of SWCNTs was qualitatively tested, as shown in Fig. 2f. After the bundle of SWCNTs were repeatedly irradiated by several flashes of light, small fires were initiated locally on the surface of the SWCNT bundle, and then slowly propagated throughout all the SWCNTs, thus confirming the potential of SWCNTs as an optical igniter.

To verify the degree of intermixing and the structure of the as-prepared SWCNT-nEM composites, we performed SEM and STEM analyses. The SEM and STEM images shown in Fig. 3a and b shows that the Al NPs were strongly connected to the surface of the CuO NPs. The SWCNTs appear to be homogeneously distributed in the Al and CuO NPs-based matrix. A STEM image (Fig. 3b) clearly shows the presence and homogeneous distribution of Al NPs, CuO NPs, and SWCNTs at a nanoscale level. This also suggests that the ultrasonication mixing process that was used with the Al/CuO/SWCNT colloidal solution in this approach was sufficiently effective.

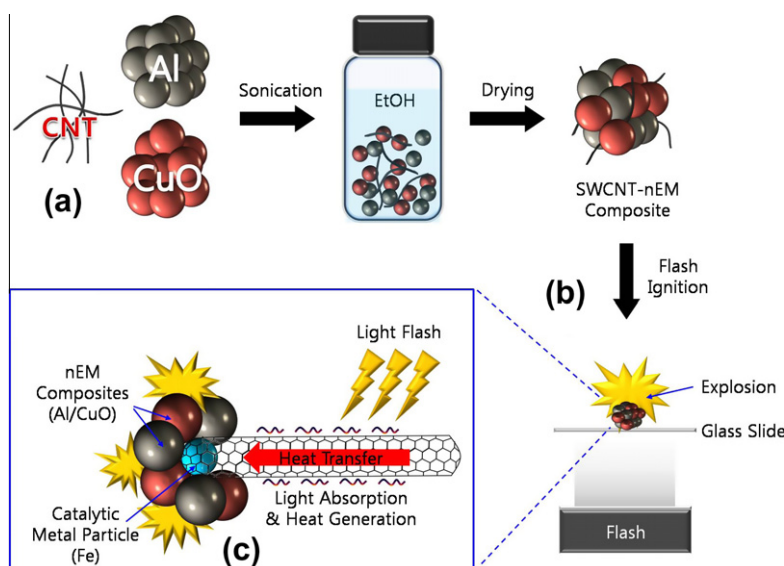


Fig. 1. Schematics of: (a) the fabrication of SWCNT-nEM composites, (b) the flash ignition of an SWCNT-nEM composite, and (c) the optical ignition mechanism of an SWCNT-nEM composite.

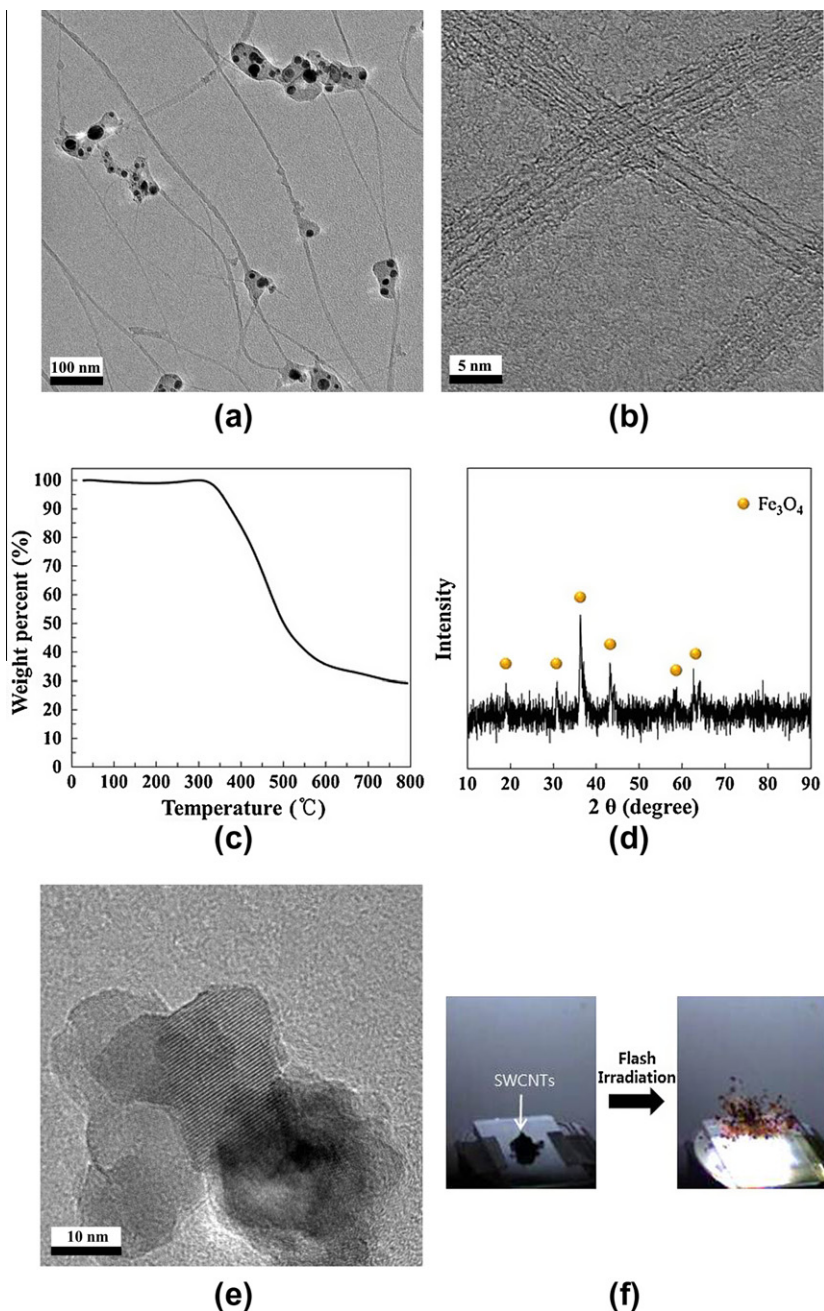


Fig. 2. (a) LRTEM and (b) HRTEM images of a bundle of SWCNTs, (c) thermogravimetric analysis of SWCNTs, (d) XRD pattern and (e) TEM image of residual materials after TG analysis (i.e., 800 °C heating) of SWCNTs, and (f) photos of a bundle of SWCNTs ignited by flash irradiation.

tive for forming a relatively homogeneous mixture of the reactants (i.e., fuel/oxidizer/optical igniter), such that the effect of the SWCNTs on the optical ignition of the nEMs could be easily recognized.

To examine the potential role of SWCNTs as an optical igniter, we performed a series of flash ignition tests on as-prepared SWCNT-nEM composite powders [1,11]. Their explosion characteristics were monitored using a high-speed camera, which was ultimately able to determine the relative differences in flame propagation speed. Fig. 4 shows a series of photographs of the flame propagation of various optically ignited SWCNT (1, 2, 5, and 10 wt.%)–nEM composite powders. Based on these photographs, the burn rates of the prepared samples were calculated. Here, the burn rate of a composite sample was determined by taking the ratio of total length of SWCNT–nEM composite powder to the time of flame propagation from one end of the SWCNT–nEM

composite powder to the other end. Fig. 4 clearly shows that the presence of SWCNTs in the nEM matrix resulted in the optical ignition of nEM composites. This took place because the temperature was raised sufficiently to initiate the oxidation of both carbon and catalysts in the SWCNT structure when the SWCNTs that were locally dispersed in the nEM matrix absorbed the irradiated flash of light (i.e., photoacoustic effect) [7]. A significant amount of heat that was generated by the carbon and the catalyst oxidation process was subsequently transferred to the neighboring nEMs for ignition and further explosion, as shown in Fig. 1c. The resulting burn rates of the SWCNT (1 wt.%)–nEM, SWCNT (2 wt.%)–nEM, SWCNT (5 wt.%)–nEM, and SWCNT (10 wt.%)–nEM composite powders were determined to be approximately 240, 180, 60, and 8 m/s, respectively. These findings suggest that increasing more than the critical amount of SWCNTs in the nEM matrix can deteriorate the exothermic reaction of the resulting SWCNT–nEM composite

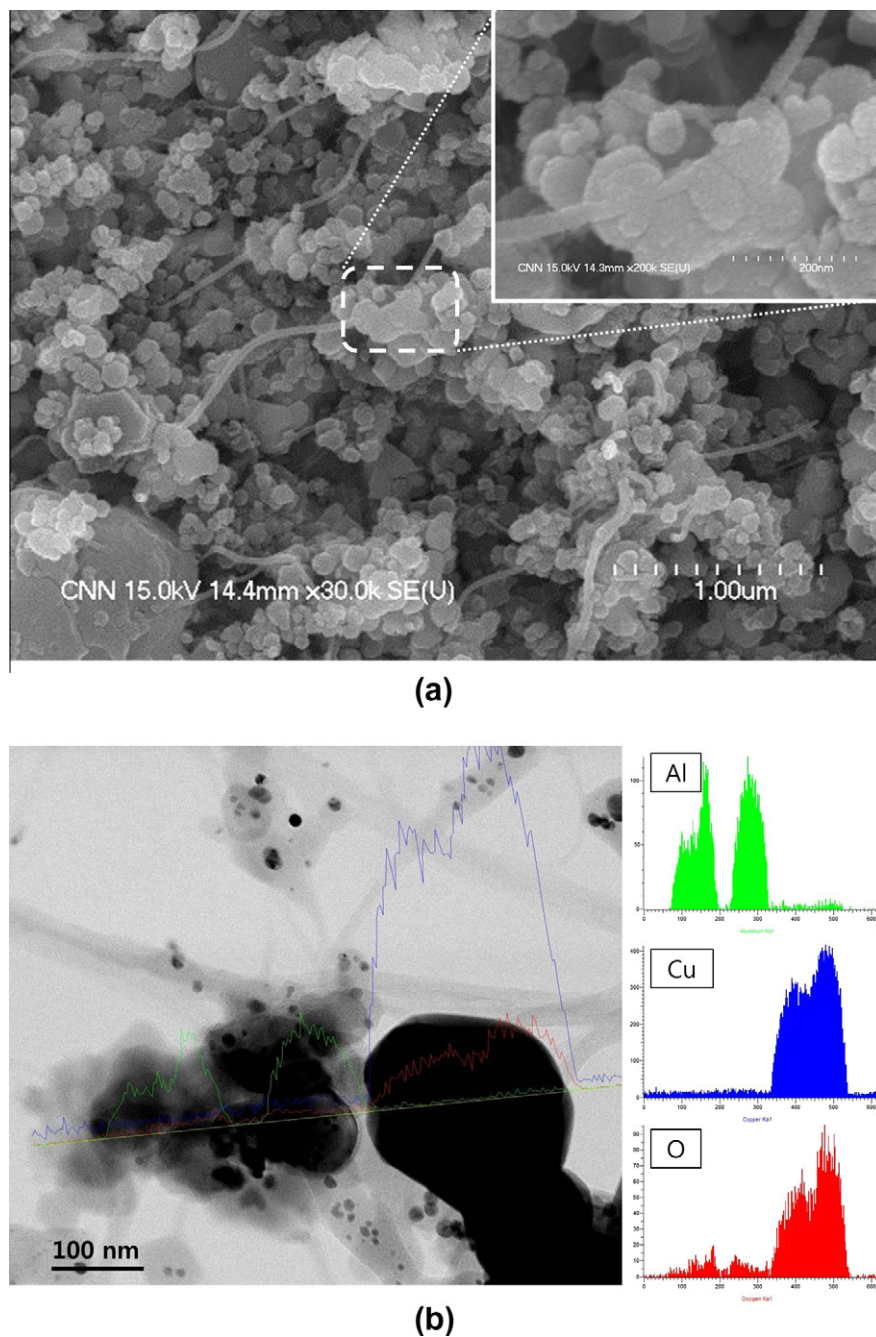


Fig. 3. (a) LRSEM image (inset: HRSEM image), and (b) scanning TEM image and line scan analysis of SWCNT/Al NP/CuO NP composites.

powders. The much slower burn rate of SWCNT (>2 wt.%)–nEM composite powders is presumably due to the fact that the presence of a large amount of SWCNTs in the given nEM matrix increased the heat dissipation through the SWCNT agglomerates, so that the temperature increase in the SWCNT–nEM composite was limited, thus hindering the oxidation and subsequent ignition of the nEMs [10,12,13]. It also should be noted that nEM powders without adding SWCNTs were able to be ignited by several repeated flashes when the light source is located close to the nEM powders. This suggests that adding an optimum amount of SWCNTs in the nEM powders can improve combustion performance with stable optical ignition.

To examine the effects of SWCNTs on the explosion reactivity of nEMs, the pressurization rate of the SWCNT–nEM composite explosion was measured by using a PCT system. A fixed amount

(~13 mg) of SWCNT–nEM composite powders was ignited in a confined cell having a constant volume of 13 mL. The pressurization rate was obtained by taking the ratio of the maximum pressure to the rise time. As shown in Fig. 5a and b, the most rapid increase in pressure and the highest pressurization rate were observed for the case of SWCNT (1 wt.%)–nEM composites. The magnitude of the maximum pressure then decreased and the rise time was reduced with an increase in the amount of SWCNT that was added (>1 wt.%). This also significantly decreased the resulting pressurization rate of the SWCNT–nEM composites, which suggests that SWCNTs play the role of a control medium for explosion reactivity, as well as that of a potential optical igniter of nEMs. By adding a relatively small amount of SWCNTs into the nEMs matrix, SWCNT–nEM composites could be effectively ignited by means of simple light flash irradiation. Simultaneously, the heat initiated

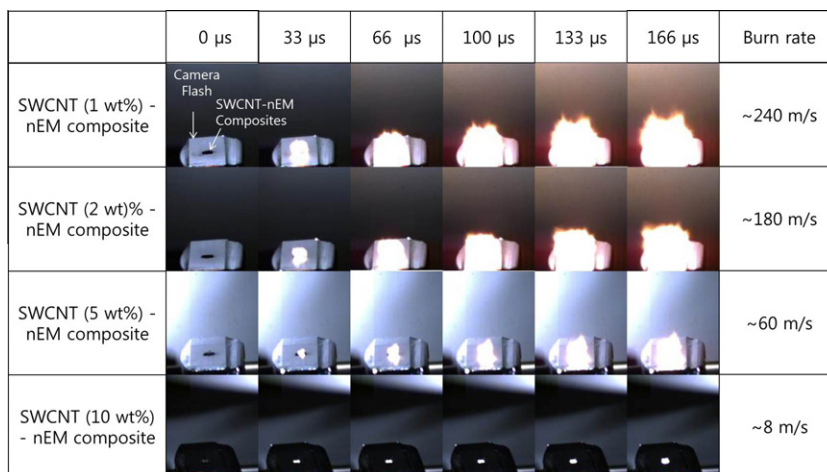


Fig. 4. High-speed camera photos of flash-ignited SWCNT-nEM composites with various amounts of SWCNTs in the nEM matrix.

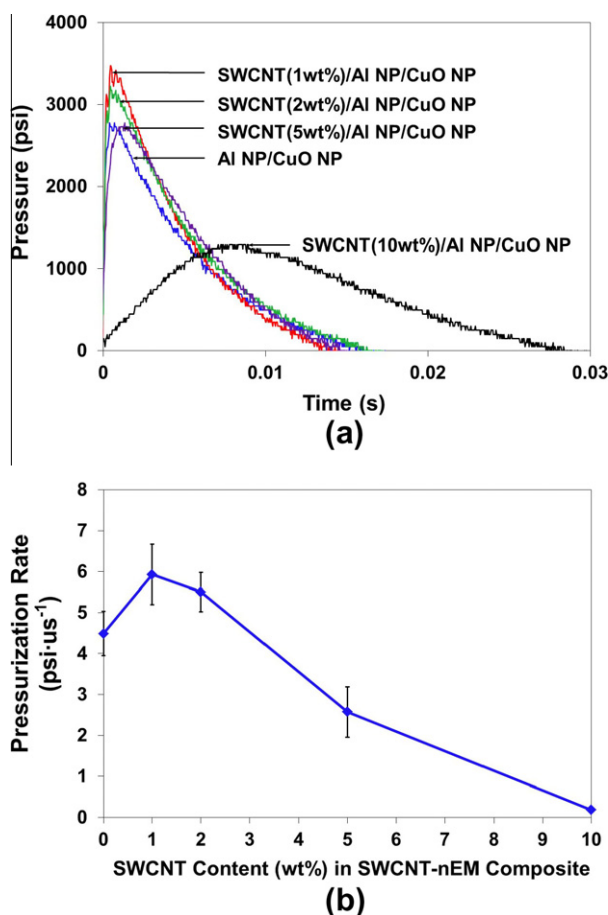


Fig. 5. Evolution of: (a) pressure traces and (b) pressurization rate of SWCNT-nEM composites as a function of SWCNT content.

by the redox reaction between Al and CuO NPs was rapidly transferred to other neighboring Al/CuO NPs through the incorporated SWCNTs, such that the resulting pressurization rate of SWCNT-nEM composites reached a maximum of $\sim 5.9 \pm 0.8$ psi/ μ s for the SWCNT (1 wt%)-nEM composite (Fig. 5b). However, the pressurization rate then decreased significantly as more SWCNTs were added into the nEM matrix (i.e., SWCNTs > 2 wt.%). This suggests that the pressurization rate of the nEMs were reduced by the corresponding increase in heat dissipation through the SWCNT

agglomeration, which eventually quenched the propagation of the explosive reaction in the nEM matrix.

4. Conclusions

We have demonstrated that the incorporation of SWCNTs in an nEM matrix makes possible the remote optical ignition of nEMs, as well as control of the explosion reactivity. Simple and easy light-flash-assisted optical ignition of nEMs was performed with various concentrations of SWCNTs (1–10 wt.%) added into the nEM matrix. However, as the SWCNT concentration in the nEM matrix increased, the corresponding increase in heat dissipation owing to SWCNT agglomeration was able to suppress the propagation of the oxidation reaction, so that the resulting burn rate and pressurization rate of the optically ignited SWCNT-nEM composites were significantly reduced. This suggests that nEMs with a desired explosive reactivity can be fabricated by incorporating a suitable amount of SWCNTs as a potential optical igniter in an nEM matrix.

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