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Different Ignition Responses of Powdery and Bulky 1,3,5-Triamino-2,4,6-trinitrobenzene (TATB) Based Polymer-bonded Explosives under Ultra-high Voltage Electrostatic Discharge

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Abstract: The electric spark induced ignition mechanism for explosives needs further study. The ignition of powdery and bulky TATB by electrostatic discharge (ESD) was investigated. Up to 200 kV ultra-high voltage ESD was applied to powdery and bulky explosives of two TATB-based polymer-bonded explosives (named PBX-1 and PBX-2). The results showed that the spark sensitivities of powdery and bulky explosives are extremely different for the same formulation. The 50% ignition voltages of powdery PBX-1 and PBX-2 were 10.8 kV and 8.5 kV, respectively, while the values for the bulky samples (tablets) were not less than 200 kV. Both heat and the electric field can be transmitted into the powdery samples, on the other hand only the electric field can be transmitted into the bulk samples. The electric field has a smaller contribution while the heat has a larger contribution to the ignition during an ESD, *i*.e., the thermal effect plays a main role in the ignition process. Our experimental results are in good agreement with recent results calculated by density functional theory.

Keywords: electrostatic spark sensitivity, ultra-high voltage electrostatic discharge, thermal effect, electric field effect

1 Introduction

Electrostatic discharge (ESD) can induce direct ignition of explosives [1]. During normal operation of explosives or ammunitions, casualties and damage may occur that are attributable to ESD induced ignition. The occurance of ESD is closely related with its energy released. Generally electrostatic sensitivity is determined by the electrostatic energy of capacitance discharge resulting in a probability of 50% initiation [2]. The electrostatic energy can be calculated from the known capacitance C (in F) of the circuit and voltage U (in V) at the condenser by means of the well-known equation $E_{ES} = 0.5CU^2$ [3]. For some sensitive explosives, even a low energy spark, of the order of 2-3 mJ, may cause initiation. The static discharge hazard is normally associated with ordinary operations, where the energy accumulated on a person may be up to 20 mJ [1].

Previous papers [4-13] have shown that the spark sensitivity of energetic materials is affected by several factors, such as chemical components, granulometry, grain shape, mechanical properties, temperature, moisture content, as well as the configuration of the electrodes and structure of the circuit. The electrode and circuit parameters correspond to the models of static discharge, such as the human body model (HBM) [14] and the helicopter model (HEM) [15].

The ESD safety of an explosive is usually evaluated by the spark sensitivity of explosive powders, such as ICM in China [16], USA organizations [17], RARDE in UK [18], Mizushima [19] and Kuroda and Nagaishi [20]. The voltage is often lower than 30 kV. For instance, the testing standard of China uses a capacitance of 30500 pF, a sample mass of 20 ±2 mg, an air gap between two electrodes (pin and board) of 0.5 mm and a highest voltage of 30 kV. This is the so-called the HBM. HBM is employed to simulate the approach of a charged human body to a detonator device or explosive. It is very commonly used within the energetic materials community [4-6].

As the air operations are more and more frequent, the HBM is no longer suitable for evaluating the ESD safety of an explosive in an actual situation. During transportation, electrostatic charge will be increasingly accumulated on the the airscrew of the helicopter, which creates a great risk to the energetic parts of the ammunition. The ESD energy on helicopters covers a wide range. Normally, the representative value can be denoted using a capacitance of 1000 pF with a voltage of 200-300 kV and a maximun discharge resistance of 1 Ω. As a result, much harsher stimulations are needed to evaluate the safety of explosives from ESD on helicopters.

Referring to the USA Military Standard (MIL-STD-331B, a Standard of the ESD test for air replenishment by helicopter), we built an installation which can implement an ESD test up to 300 kV. Using this installation, ESD tests on two TATB based polymer-bonded explosives, PBX-1 (with a formulation of 75 wt.% TATB + 20 wt.% HMX + 4 wt.% binder + other components) and PBX-2 (with a coarse formulation of 60 wt.% TATB + 35 wt.% HMX + 4 wt.% binder + other components), at ultra-high voltages were carried out. This is because these two formulations, having great potential to be employed in detonators (the most dangerous part of a warhead), have not been tested by the helicopter model spark sensitivity test. We also carried out low voltage (HBM) ESD tests on the powders of the two explosives to evaluate the safety of a charged human body to the explosive powders during daily operation. For all of the polymers and other additives used, the content of insoluble impurities was less than 0.3%. The purity of the TATB used was in the range 98.0-98.5%. TATB was synthesized in our institute by nitration of 1,3,5-trichlorobenzene to 1,3,5-trichloro-2,4,6-trinitrobenzene, followed by amination.

There are no commonly recognized ideas for an electric spark induced ignition mechanism [4-6]. Normally, larger granularities of explosives correspond to better safety performance against electrostatic discharge (ESD). Auzanneau *et al*. [7] deemed that 'hot spots' should develop at the thinnest part of the solid during ignition by ESD. These phenomena are similar to the response of an explosive undergoing thermal stimulation.

Their former explanations usually involved a thermal effect but the effect of the electric field is rarely mentioned or considered [4-6, 21]. For instance, Zeman *et al*. [4-6] found that spark sensitivity is related to the temperature, heat of combustion and thermal stability of an explosive. There are few experimental studies on the effect of the electric field, but a few simulation studies have been tried [9, 10]. The results of Cheng *et al*. [3] using the DFT-B3LYP method [22] showed that the lowest unoccupied molecular orbital energy and the Mulliken charges of the nitro group have an important effect on electric spark sensitivity. The molecular energy, dipole moment and the energy gap between HOMO and LUMO of some nitrobenzene-type explosive molecules were calculated using Density Functional Theory (DFT) by Song *et al*. [23]. It was shown that the molecular energy and the E-gap of HOMO and LUMO decreased and the dipole moment increased in an electric field. The results of Tang *et al*. [24] showed that with and without the electric field, the theoretical values are in excellent agreement with the experimental ones. Huang *et al*. [25] carried out theoretical studies on the correlation between electrostatic hazard and electronic structure for some typical primary explosives. Their study revealed that the electrostatic spark sensitivities of these primary explosives are related to their electrostatic potentials and energy gaps. Despite these successful researches, the role of the external electric field in ignition by ESD is not yet well understood.

Generally, during an ESD, the explosive is stimulated by the electric field and high temperature [26, 27]. The electric field polarizes the explosive molecule [23] and the thermal effect may break chemical bonds to initiate decomposition. In the present work, electric field effects as well as the thermal effect were considered in studying the safety performance of PBX-1 and PBX-2.

2 Experimental

2.1 Test method for explosive powders

The test arrangement for the electrostatic spark method for explosive powders is shown in Figure 1. The discharging capacitance was 30500 pF. The air gap between the electrodes was set at 0.5 mm. The explosive powder was stimulated by the electrostatic spark during an ESD through the air gap. The mass of sample was set at 20 ±2 mg. The sample of 20 mg was rolled out on the board electrode and the top of the polymethyl methacrylate (PMMA) sleeve was covered with transparent adhesive tape, thus a closed cavity was formed inside the PMMA sleeve. The transparent adhesive tape was then pierced with a pin electrode, setting the air gap between the two electrodes at 0.5 mm. The pin electrode was the negatively charged electrode, namely a cathode (−), in our test (Figure 1). The up-and-down procedure (UDP) [28] was applied in selecting the voltage of every attempt during the ESD tests for the samples of PBX-1 and PBX-2. The higest voltage applied to the explosive powders was 30 kV.

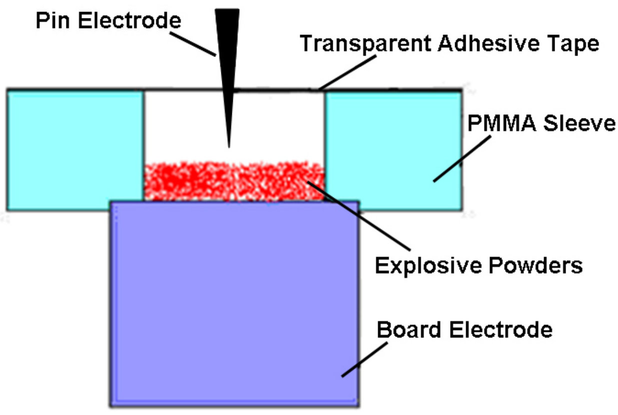


Figure 1. Illustration of the electrostatic spark test method for explosive powders

2.2 Test method for explosive tablets

The test arrangement of the electrostatic spark method for explosive tablets is shown in Figure 2. The air gap between the electrodes was set at 1.5 mm. The mass of the sample was set at 0.7 g with a diameter 10mm and thickness 5 mm. The sample was positioned on a PMMA base, and two parallel pin electrodes with a separation of 1.5 mm were fixed on the surface of the tablet by a holder made of PMMA. The ESD of the Pin-Pin discharge was employed. The explosive tablet was stimulated by the electrostatic spark during the ESD (Figure 2). The method of explosion probability was applied for different voltages. The ultra-high voltage electrostatic discharge equipment used in this work is shown in Figure 3. The test was initiated at 60 kV and an increment of 20 kV was added if no reaction occurred in 7 ESD at each voltage until reaction occurred or until the voltage reached 200 kV.

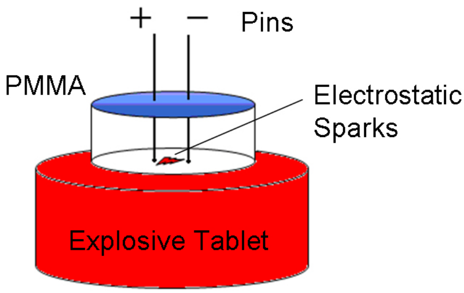


Figure 2. Illustration of the electrostatic spark test method for explosive tablets

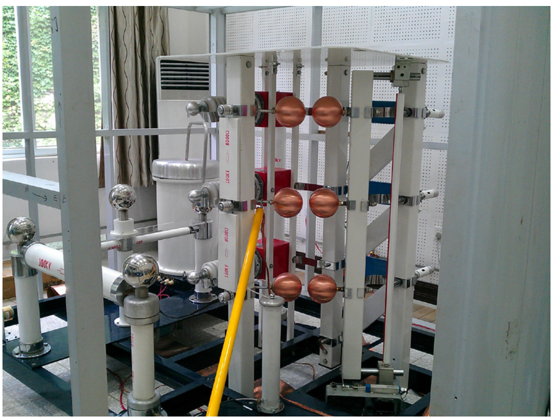


Figure 3. Ultra-high voltage electrostatic discharge equipment

The range of voltages the equipment could discharge was between 0 kV and 300 kV. The discharging capacitance was 1000 pF and the discharging resistance was 1 Ω, with a discharging inductance of 20 μH. These technical details obey the American Military Standard (MIL-STD-331B), and this model is called the 'Helicopter Model'.

To identify an appropriate value for the distance of the air gap between two pin-electrodes, ESD tests were performed under various air gaps to investigate the effect of the air gap distance on the discharging voltage threshold. As mentioned earlier, the air gap distance was finally set at 1.5 mm; the experimental data is presented here as support for this value.

It should be mentioned that, the reaction degrees under ESD included no ignition or ignition, and then extinguished, combustion, deflagration and detonation. For an explosive tablet, if the surface turned black but the tablet kept its shape and no further reaction occurred, it was defined as no ignition.

3 Test results

3.1 Test results of the explosive powders

The ESD results of PBX-1 and PBX-2 powders are listed in Table 1.

Table 1. Test results for PBX-1 and PBX-2 powders

Sample	Static voltage V_{50}* [kV]	Standard deviation [kV]	Static energy E_{50} [J]	Temperature [°C]	Relative humidity [%]
PBX-1	10.83	2.03	1.789	16	40
PBX-2	8.54	1.03	1.112	16	50

*V_{50} is the stimulus voltage at which 50% of the tests explode.

The V_{50} values for PBX-1 and PBX-2 powders were 10.83 kV and 8.54 kV, respectively. In other words, under a stimulus of an ESD of 10.83 kV and 8.54 kV, 50% of the samples of PBX-1 and PBX-2 exploded, respectively. The corresponding energies (E_{50}) were 1.789 J and 1.112 J, respectively. After reaction, a lingering smell of burning was observed. Partial or complete consumption of the explosive sample occurred. The 50% initiation probability of a TATB based explosive is much lower than common explosives or conventional propellants, *e.g.* the V_{50} of ANPs and aluminum micron powders are 1.42 kV and 1.53 kV [29].

3.2 Test results of ESD with various air gaps

To study the effect of various air gaps on ESD and to help to choose the best distance between two electrodes, we implemented a series of ESD tests in which the air gap between the two electrodes was 0.5 mm, 1.5 mm and 4.5 mm. The breakdown voltages at different air gap thicknesses were measured according to the method of Jin *et al*. [30]. A discharging capacitor of 500 pF and a discharging resistor of 100 Ω were used. We tuned the voltage to a specific value and then turned on the discharging switch. Every specific value of the voltage was discharged ten times, after which we calculated the flash probabilities. When the discharge occurred, we saw a spark or flash. The spark or flash is a sign of ESD. For every value of the air gap and every voltage 10 attempts were performed. ESD stimuli were judged to be stable provided that ESD occurred in all 10 attempts. The temperature was 18 °C and the air humidity was 56% RH when the tests were performed. The tests results are listed in Table 2.

Table 2. Flash probabilities with various air gaps (10 attempts for each point, 18 °C, 56% RH)

<table>
<tr><th>Voltage [kV]</th><th>Flash probabilities under 0.5 mm air gap [%]</th><th>Voltage [kV]</th><th>Flash probabilities under 1.5 mm air gap [%]</th><th>Voltage [kV]</th><th>Flash probabilities under 4.5 mm air gap [%]</th></tr>
<tr><td>1.9</td><td>0</td><td>2.5</td><td>0</td><td>6.4</td><td>0</td></tr>
<tr><td>2.0</td><td>20</td><td>2.6</td><td>60</td><td>6.6</td><td>10</td></tr>
<tr><td>2.1</td><td>30</td><td>2.7</td><td>90</td><td>6.8</td><td>10</td></tr>
<tr><td>2.2</td><td>40</td><td>2.8</td><td>100</td><td>7.0</td><td>10</td></tr>
<tr><td>2.3</td><td>60</td><td>-</td><td>-</td><td>7.2</td><td>20</td></tr>
<tr><td>2.4</td><td>100</td><td>-</td><td>-</td><td>7.4</td><td>30</td></tr>
<tr><td colspan="4" rowspan="4">The bandwidth of voltages was the narrowest from 0% to 100% flash probabilities (2.5-2.8 kV) for the 1.5 mm air gap. Thus this is the best choice for the pin separation.</td><td>7.6</td><td>40</td></tr>
<tr><td>7.8</td><td>70</td></tr>
<tr><td>8.0</td><td>90</td></tr>
<tr><td>8.2</td><td>100</td></tr>
</table>

From Table 2 one can see that, at 18 °C and 56% RH, for a 0.5 mm air gap between the two pins, the bandwidth of the breakdown voltage threshold was between 1.9 kV and 2.4 kV. Thus, voltages below or equal to 1.9 kV cannot cause an ESD, voltages between 1.9 kV and 2.3 kV may cause an ESD, and voltages above or equal to 2.4 kV can definitely cause an ESD. For a 1.5 mm air gap, the bandwidth was between 2.5 kV and 2.8 kV. For a 4.5 mm air gap, the bandwidth was between 6.4 kV and 8.2 kV. According to the results in

Table 2, the threshold of the spark breakdown voltages are proportional to the air gap between the electrodes (Figure 4). When the air gap was increased, the breakdown voltage was enhanced. The bandwidth of voltages was the narrowest, from 0% to 100% flash probabilities (2.5-2.8 kV), for a 1.5 mm air gap (Figure 4). A narrow bandwidth of voltages is beneficial for higher reproducibility of the tests. So 1.5 mm is the best choice for the pin separation. Consequently, we set the air gap at 1.5 mm for the tablet ESD tests.

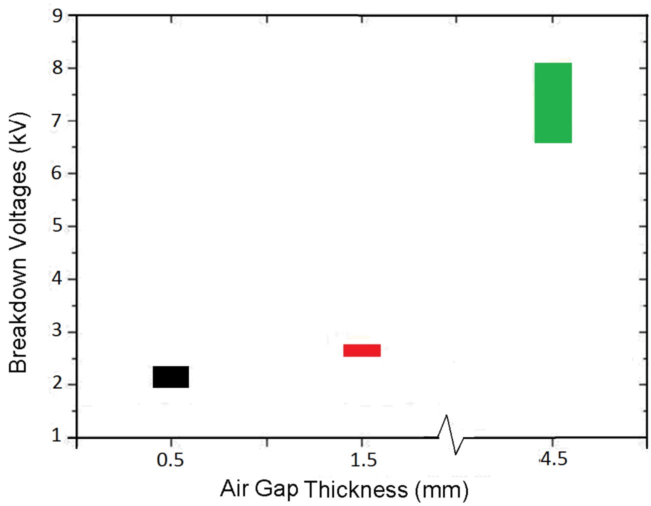


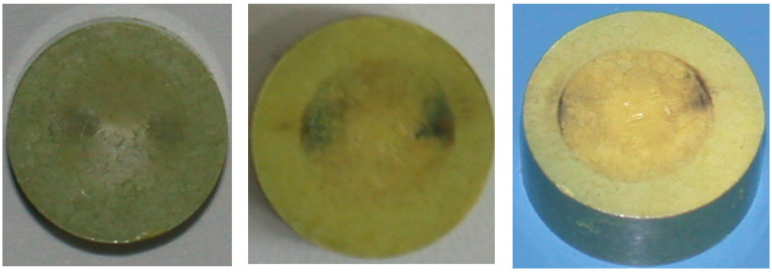
Figure 4. Breakdown voltage *vs*. air gap thickness, at 18 °C and 56% RH

3.3 Test results for explosive tablets

3.3.1 Test results for PBX-1 tablets

The pictures of PBX-1 samples after a wide range of high voltages stimuli are shown in Figure 5. These show that after ultra-high voltage ESD, no self-sustaining combustions or explosions occurred for PBX-1 tablets.

For PBX-1 tablets, 25 attempts of the test at every 20 kV voltage from 60 kV to 200 kV were performed. The reaction probabilities for the tablets were all zero under a wide range of high voltage stimuli, indicating that the tablets of PBX-1 possess a very good capacity to resist ultra-high voltage electrostatic discharge, better than PBX-1 powder.

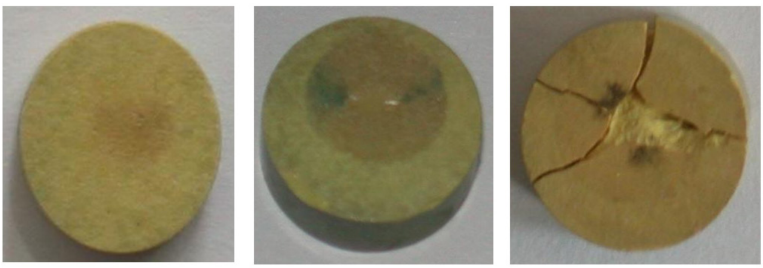


After 7 EDSs at 60 kV After 7 EDSs at 120 kV After 7 EDSs at 200 kV

Figure 5. Tablets of PBX-1 after ESD at various voltages

3.3.2 Test results for PBX-2 tablets

The pictures of PBX-2 samples after a wide range of high voltages stimuli are shown in Figure 6. These show that after ultra-high voltage ESDs, no self-sustaining combustions or explosions occurred for PBX-2 tablets. The tablets cracked under the pressure of sparks, such as the short impulse stress of an ESD [31, 32] of 200 kV. Although the stimulating voltages were very high and despite the tablet being torn into small pieces by the small shock waves produced by the sparks, still no combustion and ignition occured.



After 7 EDSs at 60 kV After 7 EDSs at 120 kV After 7 EDSs at 200 kV

Figure 6. Tablets of PBX-2 after ESD at various voltages

In the test for PBX-2 tablets at eight high voltage ESDs, from 60 kV to 200 kV, the reaction probabilities were all zero, indicating that the tablets of PBX-2 possess a very good capacity to resist ultra-high voltage electrostatic discharge, better than explosive powder of the same formulation.

In Figures 5 and 6, the black spots on the surfaces of the tablets are the trails of decompositions. During the sparks, high temperature as well as air shockwaves (a consequence of the inflation of air heated by the sparks) occurred. The explosive on the surface may be shocked, peeled off and decomposed at higher temperature. The reaction cannot self-sustain to burning or explosion. As a result, black spots are left on the surfaces of the tablets as trails of decomposition. Thus, the ESD effects on tablets are the combined effects of shockwave loading and thermal heating.

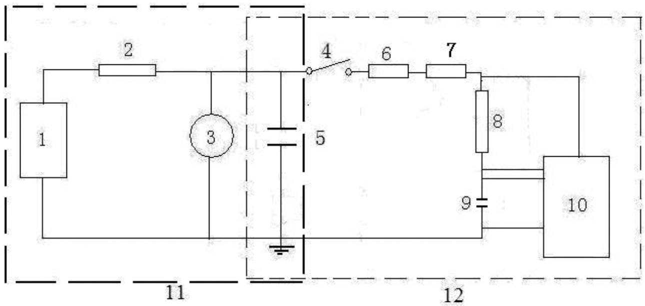
4 Analysis and Discussion

The test results show that PBX in a tablet form is very tough during an ESD. In all of the hundreds of ESD tests, none of the tablets were ignited, even at the ultra-high voltage of 200 kV and repeated 7 times. Figure 6 shows the tablet after these rigorous tests. No ignition was observed even though the ϕ 10 mm × 5 mm PBX-2 tablet was torn into fragments by the shockwaves of the spark.

The capacitance discharging spark ignition mechanism can be summarized as follows. The air between the electrodes is ionized and penetrated by the instantaneous strong static electric field. As a result, a huge electric current forms and a flash of light is emitted. A local high temperature spark is the main reason for ignition of the explosive. 5 m long air discharges were created and the temperatures of the sparks were measured. The temperatures were in the range of 2.0×10^4 K to 3.4×10^4 K. Experiments were also performed on a spark of 2.5 m length and the temperature was identical to the values given above [26]. The temperatures of the N_2 molecules were also measured in the capacitance spark discharge. The discharge voltage was 3.0-3.5 kV and the discharge energy was 0.03-1 mJ. The rotational and vibrational temperatures of N_2 are estimated to be 500 K and 5000 K, respectively [27].

On the whole, during an ESD the explosive is stimulated by the electric field and high temperature. The electric field polarizes the explosive molecules and the thermal effect cleaves the chemical bonds to initiate decomposition. Whether the electric field effect and the thermal effect occur in the same time frame is still not clear. We called this ‘the asynchrony problem’.

To investigate this asynchrony problem, we have initially to find the parameters of the electric field effect and thermal effect. Secondly, the time interval for these parameters has to be determined. According to the spark ignition mechanism, the voltage across the gap was chosen as the parameter to denote the electric field effect, and the electric current across the gap was chosen as the parameter to denote the thermal effect. The history of an ESD was studied by measuring the waveforms of voltage and current across the gap in the electric circuit shown in Figure 7. The methods for measuring the ESD wave forms were described by Lin [33].



1 − direct current high voltage electrical source (ESD-30); 2 − charging resistance (100 Ω); 3 − static voltage meter; 4 − high voltage switch; 5 − capacitance, 500 pF; 6 and 7 − discharging resistances (160 Ω); 8 − sampling resistance (80 Ω); 9 − ESD gap (1.5 mm); 10 − data collecting system (oscillograph); 11 − charging part; 12 − discharging part
Figure 7. The discharging electrical circuit

The signal of the gap voltage connected to the oscillograph, after being attenuated 1000X, and then the waveforms of the voltage were obtained. The signal of the voltage of the sampling resistance connected to the oscillograph, after being attenuated 1000X, was then divided by the value of the resistance (80 Ω). The result was the waveform of the current across the gap.

The electrical source voltages were adjusted to 7 kV, 8 kV, 9 kV and 10 kV to test the response of the circuit. The peak values of voltage and current and their time intervals were recorded or calculated, and are listed in Table 3. The shapes of the waveforms were similar, despite the differences in the electrical source voltages, except for their peak values.

Table 3. Peak values of voltage and current and their time intervals

Capacitance voltage [kV]	Gap voltage [kV]	Gap current [A]	Time interval [ns]
7	6.96	44.1	23.4
8	7.86	50.5	24.4
9	8.39	53.7	25.6
10	9.36	64.1	24.8

As shown in Table 3, the peak value of the gap voltage is close to the capacitance voltage.

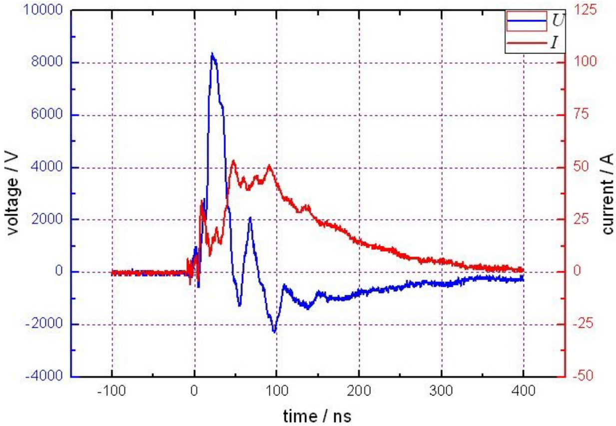


Figure 8. The waveforms of voltage and current across the gap at an ESD of 9 kV

The waveforms of voltage and current across the gap at an ESD of 9 kV are shown in Figure 8. This indicates that there is a time interval between the peak of the gap voltage and the peak of the gap current. The peak of the gap voltage was ~25 ns ahead of the peak of the gap current. When the gap current reaches its peak, the gap voltage has decreased to almost zero, indicating that there is asynchrony in the thermal effect and the electric field effect during an ESD spark. This asynchrony of the electric field effect and the thermal effect during an ESD is the time interval between the peak of the electric field effect and the peak of the thermal effect. The electric field effect is ahead of the thermal effect. This time interval weakens the effect of an ESD on an explosive, which causes the ESD to exhibit macroscopically the main effect of the spark, namely the thermal effect.

The electric field can pass through both the powders and the tablets. On the other hand, the spark can easily pass through the powders but not the tablet. The heat transfered and absorbed are involved in the thermal effect. Hence the surface area is very important for the response of the thermal stimuli. The grain scale of the powder samples of PBX-1 and PBX-2 (with a V50 of about 10 kV) was about 40 μm, so the specific surface area was about 75 mm^{-1}. Bulk samples like the ϕ 10 mm × 5 mm tablets (no ignition at 200 kV ESD) has a specific surface area of 0.6 mm^{-1}, which is much smaller than the value of the powder samples. Here the smaller surface area corresponds with a much higher V50. The correlation between V50 and the surface area also indicates that the ESD is macroscopically mainly a thermal effect.

The molecular energy, dipole moments and energy of the gaps (E_{gap}) of HOMO and LUMO of some nitrobenzene explosive molecules were calculated using Density Functional Theory by Song *et al*. [23]. It was shown that the molecular energy and the E_{gap} of HOMO and LUMO decreases and the dipole moment increases in an electric field. These results indicate a near linear correlation between the electric spark sensitivity of explosives and the E_{gap} of the molecules, namely, a smaller E_{gap} corresponds with a lower electric spark sensitivity. A decrease in molecular energy indicates a more stable molecular structure in an external electric field, which is consistent with our explanation of our experimental observations.

5 Conclusions

The ignition of powdery and bulky TATB by an electrostatic discharge (ESD) was investigated. Ultra-high voltage ESD tests were carried out on both powdery and bulky explosives of TATB-based polymer-bonded explosives. The results showed that the spark sensitivities of powdery and bulky explosives are extremely different for even the same formulation. There were strong correlations between the spark sensitivities and the states of the explosives. The 50% ignition voltages of powdery PBX-1 and PBX-2 were 10.8 kV and 8.5 kV respectively, while the values for the bulky ones were not less than 200 kV. In order to explain this observation, the contributions of heat and electric field were studied by measuring the spark current and voltage waveforms of the circuit in real time. The results showed that the loadings of spark current and voltage cannot be synchronized across a gap, indicating there is a time interval between the thermal effect and the electric field effect. After the peak of the voltage has passed, the spark current gradually increases. There is a time interval of ~25 ns between the two peaks, greater than the molecular vibration periods (of the order of 10 ps) according to molecular vibration theory [34]. Combined with the available literature [23], we can draw a conclusion that the molecules of explosives are polarized during the loading of an external electric field. However these polarized molecules will be rapidly restored to their original structure, in a time such as 10 ps. Both the heat and electric field can transmit deep into the PBX powers; on the other hand only the electric field can transmit deep into the bulk. Thus the electric field has a smaller contribution while the heat has a larger contribution to the ignition during an ESD, that is the thermal effect plays a main role in the ignition process. The time interval weakens the effect of an ESD on an explosive, which causes the ESD to exhibit macroscopically the main effect of thermal sparks. Our experimental results are in good agreement with recent DFT calculated results. Our results provide academic insight and experimental evidence for electrostatic safety management in explosives manufacture and applications.

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