Research Progress of Effects on Nanomaterials on Pyrotechnic Properties

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Abstract. Nanomaterials have been widely concerned by researchers in recent years, because of its unique physical and chemical properties, which play an irreplaceable role in improving the performance and safety of pyrotechnics. In this paper, in the light of the effects of nanomaterials on pyrotechnic properties such as metal nanoparticles, metal oxide nanoparticles, composite nanoparticles, carbon-based nanomaterials, the latest research progress and current research situation are reviewed from two aspects including thermal decomposition temperature and decomposition heat, and the function mechanism was analysed. The future research directions are expected.

1. Introduction
Pyrotechnics is composed of combustible agent, oxidant, binder and functional additive, which can react quickly and produce many effects such as sound, light, smoke and thermal[1-2]. It is widely used in civil and military fields, but there are some problems, such as waste of resources, unsatisfactory effect, low safety factor during use and storage, which are caused by insufficient reaction of reagents. Therefore, it poses a severe challenge to the development and application of pyrotechnics.

Fig. 1 Application of nano-materials and technology in pyrotechnics
In recent years, the rapid development of research and development and preparation technology of new nano-materials has gradually solved these problems. The specific application in pyrotechnics is
shown in Figure 1. In this paper, the effects of nano-materials on thermal decomposition temperature and decomposition thermal of pyrotechnics are reviewed, which provides reference and support for the next pyrotechnics formula optimization.

2. Influence on thermal decomposition temperature of pyrotechnics

2.1. Metal nanoparticles
Aluminum powder is a kind of combustible agent often used in pyrotechnics, and its nanoparticles can greatly reduce the activation energy of oxidant thermal decomposition, thus reducing its thermal decomposition temperature. Zhang et al. found that the initial reaction temperature of pyrotechnic agent system containing nano-aluminum powder (50 nm) decreased by 3.8%, which indicated that the difficulty of thermal reaction of pyrotechnic agent added nano-aluminum powder was effectively improved. Pantoya et al. found that the ignition temperature of thermit containing nano-Al powder (17–202 nm) mixed by mechanical means decreased significantly, which was mainly due to the decrease of nano-material size, which led to a sharp increase in the specific surface area of components, and the contact between combustible agent and oxidant was more sufficient, which effectively reduced the diffusion and reaction distance, thus reducing the reaction temperature.

At the same time, further study found that the initial decomposition temperature of the ultra-refined KClO₃ and Al system (KClO₃ is submicron, about 300 nm, Al powder is nanometer, 50 nm) is about 14°C lower than that of the samples before refinement. The reason is that the surface active position of particles in ultrafine pyrotechnics increases, and the chemical reaction activity increases. The thermal generated in the mixed system is not easy to dissipate, and the thermal can be absorbed quickly to reach the activation temperature required by the reaction.

In addition, Tan Linghua et al. found that nano-Cu particles also have significant catalytic effect on the thermal decomposition of NH₄ClO₄ (AP). The exothermic peak temperature of AP at low temperature and high temperature decreased by 15°C and 17.6°C respectively with 26 μm copper powder, respectively, while that of nano-Cu (20 nm) was 35.1°C and 130.2°C, and the decrease of exothermic peak temperature of AP increased with the decrease of Cu powder particle size.

2.2. Metal oxide nanoparticles
Metal oxide nanoparticles play an important role in catalysis and energy conversion applications of nanotechnology. Chen et al. found that nano-Co₃O₄ particles can reduce the peak temperature of AP at high temperature by 128.5°C, and the smaller the particle size, the more the thermal decomposition reaction temperature of AP decreases. Ma et al. explained by analysis and comparative study that micron Fe₂O₃ made the first and second exothermic peaks of AP move to low temperature by 1.10°C and 62.25°C, respectively, while the values of nano-Fe₂O₃ were 61.89°C and 118.82°C, which indicates that the catalytic activity of nano-Fe₂O₃ was obviously better than that of micron Fe₂O₃. Zhang et al. found that nano-CoFe₂O₄, NiFe₂O₄ and ZnFe₂O₄ reduced the high temperature decomposition peak temperature of AP by 108.99°C, 82.03°C and 30.91°C, respectively, among which CoFe₂O₄ played the most obvious role because the interaction between iron and cobalt was beneficial to the thermal decomposition of AP, but the main reason was that smaller microcrystals and particle sizes contained more lattice defects and active sites, which improved the reactivity and thus reduced the decomposition temperature of AP.

In order to explore the influence of the preparation method of nanomaterials on the thermal decomposition performance of AP, Hao Gazi et al. found through experiments that the catalytic effect of nano-CuCr₂O₄ prepared by mechanical crushing method is more obvious than that prepared by coprecipitation method. Figure 2 and 3 are TEM images of nano-CuCr₂O₄ directly prepared by coprecipitation method and TEM diagrams of nano-CuCr₂O₄ prepared by grinding method, respectively. It can be seen that the particle size of nano-CuCr₂O₄ prepared by coprecipitation method is about 30 nm, and its distribution is uniform, but its dispersion is poor. The particle size of CuCr₂O₄ prepared by mechanical pulverization is about 60 nm, and the dispersion of CuCr₂O₄ is better. Zhu et
al. [12] found that the average particle size of spherical nano-CuO samples obtained by adding NaOH at 100°C is the smallest (about 6 nm), which reduces the decomposition temperature of AP by 10°C, which is better than 96°C of needle-like nano-CuO, because spherical nano-CuO has larger specific surface area and stronger catalytic activity.

2.3. Composite nanoparticles
By preparing nanocomposite materials of nanoparticles and reactants, the reaction distance can be greatly reduced, the contact area can be increased, and the catalytic effect can be improved. Song et al. [13] studied that the high and low temperature decomposition peaks of AP/Fe₂O₃ nano-composite oxidant prepared by sol-gel method were 124.96°C and 113.02°C earlier than those of pure AP, respectively. This is because the low valence oxides on the surface of composite particles play a bridge role. The transfer of electrons from ClO₄⁻ to NH₄⁺ is promoted, and the specific surface area of nano-Fe₂O₃ increases, which increases the proportion of surface atoms. A large number of unsaturated suspended bonds and unsaturated bonds, a large number of active points and lattice defects on the surface would adsorb a large number of gas phase decomposition products of AP, which significantly promotes the high temperature decomposition of AP.

At the same time, composite nano-materials can obviously solve the agglomeration problem of nano-particles. Chen et al. [14] prepared CuO/AP composite particles with nano-CuO as the core and AP as the shell by solvent-non-solvent method, which effectively solved the problem that nano-CuO particles are easy to agglomerate, and at the same time, it advanced the high temperature decomposition peak temperature of AP by 101°C.

2.4. Carbon-based nanoparticles
Carbon is the most widely distributed element in nature. Zero-dimensional, one-dimensional, two-dimensional and three-dimensional carbon nanomaterials based on carbon can be widely used as carriers of energetic components by coating or packaging because of their unique physical structure and chemical properties, as well as their green and pollution-free characteristics [15].

The open pore structure of carbon nanotubes (CNTs) is beneficial to electron transfer and thermal conduction. At the same time, loading catalyst particles on the wall of CNTs can effectively prevent the agglomeration of nanoparticles. Compared with simple mixed samples, CNTs and Fe₂O₃ composite particles prepared by Jiang et al. [16] can further reduce the peak temperature of AP pyrolysis and have better catalytic performance. Meng et al. [17] also deposited titanium dioxide coating with a thickness of about 2 nm on CNTs surface by sol-gel method. As seen in Figure 4, the composite can continuously reduce the peak temperature of AP pyrolysis by 52.5°C, which indicates that the composite can further improve the catalytic performance of TiO₂ particles as AP thermal decomposition carriers. Metal Ag/CNTs nanoparticles also catalyzed the pyrotechnics of magnesium/potassium nitrate system, and the exothermic peak of ignition reaction of pyrotechnics decreased by 37°C [18]. It is found that the thermal decomposition temperature would also be reduced and the reaction rate would be improved by combining CNTs with oxidant particles to prepare composite particles. Because it avoids the agglomeration of nano-scale oxidant particles, it would...
greatly reduce the particle size of oxidant loaded on CNTs, and improve the endothermic and exothermic rates of the reaction.

Compared with carbon nanotubes, graphene (GA) has an ultra-thin nanosheet structure, which means that it has a higher specific surface area and can support more catalysts. Li et al. [30] found that nano-MnO₂-GR composite reduced the thermal decomposition temperature of AP by 141.9 °C. In AP/GA aerogel nanocomposites (AP average particle size is about 69.41 nm) prepared by Wang Baoxue et al. [21], the peak temperature of AP pyrolysis decreased by 83.7 °C. The analysis shows that: On the one hand, graphene has a large specific surface area, which can adsorb NH₃ and HClO₄ to the surface when AP is decomposed at low temperature, delay their entry into gas phase and oxidation reaction, and then greatly weaken the decomposition of AP at low temperature; On the other hand, the decomposition product of HClO₄ reacts with graphene, which gives off a lot of thermal and causes the surface temperature of AP crystal to increase. As a result, pyrolysis occurs ahead of time.

In conclusion, the application of nano-materials can significantly reduce the thermal decomposition temperature of pyrotechnics. Compared with the nanocrystallization of pyrotechnics components, nano-catalysts and their composite materials, the addition of carbon nano-materials can significantly reduce the thermal decomposition temperature, as shown in Table 1.

| Nanomaterial Classification | Nanoparticles | Particle Size/ nm | Temperature Drop Range/°C | Reduction Range | Researchers |
|-----------------------------|---------------|-------------------|---------------------------|----------------|-------------|
| Metal Nanoparticles         | Al            | 50 nm±             | 4.63                      | 3.8%           | Zhang, et al. [37] |
|                             | Al sub-micron KClO₃ | KClO₃: 300 nm±, Al50 nm± | 14| 11.3% | Zhang, et al. [39] |
|                             | Cu            | 20 nm±             | 130.2                     | 27.3%          | Tan, et al. [6] |
|                             | Co₃O₄         | 21.8 nm±           | 128.5                     | 28.4%          | Chen, et al. [14] |
| Metal Oxide Nanoparticles   | FeO₂          | 8-25 nm±           | 118.82                    | 25.3%          | Ma, et al. [9] |
|                             | CoFe₂O₄       | 220 nm±            | 108.99                    | 26.9%          | Zhang, et al. [10] |
|                             | CuO           | 19-68 nm           | 103                       | 22.9%          | Zhu, et al. [12] |
| Composite Nanoparticles     | AP/FeO₂       | AP 19-68 nm        | 124.96                    | 28.7%          | Song, et al. [13] |
|                             | Nano-CuO/AP   | CuO 40 nm±         | 101                       | 22.3%          | Chen, et al. [14] |
|                             | Fe₃O₄/CNTs    | Fe₃O₄: 10-30 nm    | 122.1                     | 25.5%          | Jiang, et al. [18] |
| Carbon-based Nanomaterials  | Ag/CNTs       | Ag 69.41 nm±       | 37.1                      | 5.9%           | Pourretadl, et al. [18] |
|                             | TiO₂/CNTs     | TiO₂: 2 nm±        | 52.5                      | 12.3%          | Zhang, et al. [19] |
|                             | MnO₂-GR       | MnO₂:10 nm±        | 141.9                     | 32.7%          | Li, et al. [20] |
|                             | AP/GA         | AP 69.41 nm        | 83.7                      | 20.6%          | Wang, et al. [21] |

3. Influence on decomposition thermal of pyrotechnics

3.1. Metal nanoparticles
Tan et al. [6] found that when copper powder reaches nanometer level (20 nm), the apparent decomposition thermal of AP pyrolysis can be increased to 1.20 kJ/g, which is 172.7% higher than
that of commercial micron copper powder (26 μm) with the same dose. This is due to the large specific surface area of nano-copper powder, which can promote the chemical reaction and thermal release between gases, greatly promote the decomposition thermal of high temperature decomposition. Zhang et al. [22] found that in Zr/KClO₄, the traditional formula of pyrotechnic laser pump source, with the decrease of Zr particle size, the radiation energy increased obviously, and the peak radiation intensity doubled, which greatly increased the laser output energy.

3.2. Metal oxides and their composite nanoparticles
Ma et al. [9] found that the constant volume combustion thermal of simulated solid propellant increased by 5095.70 J/g and 2350.84 J/g respectively compared with pure AP after adding nano-Fe₂O₃ and micron Fe₂O₃, and the effect was remarkable. The reduction of AP particle size also contributed to the improvement of propellant decomposition thermal[23]. Wang et al. [24] found that the total thermal release of Fe₂O₃/Al nanocomposites prepared by sol-gel method reached 1648 J/g, which was 95.7% higher than that of micron-sized mixtures, and showed better ignition and energy characteristics than traditional materials. Sherif et al. [25] prepared TiO₂ colloidal particles with an average particle size of 10 nm by hydrothermal method. It was found that TiO₂ combined the two main thermal release stages of AP and increased the total thermal release by 18%. Wu et al. [26] successfully prepared a CuO/Al nano-composite energetic material with more uniform microstructure by DNA self-assembled method. Figure 5 is the TEM and SEM images of physically mixed CuO/Al samples and DNA self-assembled. It can be found that most CuO and Al in the physical blend samples (Figure 5a and 5c) are aggregated and only a few are in close contact, and DNA self-assembled samples (Figure 5b and 5d). The distribution of CuO and Al in the samples was uniform, and there was no obvious aggregation of nanoparticles. The experimental results show that the reaction thermal of DNA self-assembled samples is higher than that of physical blend samples with the same ratio, and the maximum increase range can reach 52.15%. The nanocrystallization of Al nanoparticles is also an important factor leading to the obvious improvement of thermal reaction efficiency and rate.

3.3. Carbon-based nanomaterials
Sherif et al. [15] studied the catalytic effect of reduced graphene oxide (RGO) and Fe₂O₃ nanocomposites on AP, and found that the total thermal release of AP increased by 83%. The main
reason is that the superior interface surface area of RGO ensures the absorption of gas products on the catalyst surface, and nano-powder can reduce the required endothermic phase transition, and finally make the total thermal released surge. Lan et al. [27] found that compared with the decomposition thermal of pure AN of 1048. 62 J/g, the decomposition thermal of mechanical mixture of GA and AN increased by 251.9 J/g. Graphene aerogel/ammonium nitrate (GA/AN) nanocomposites (AN average particle size 71 nm) were prepared by sol-gel method and supercritical CO$_2$ drying method. As can be seen from Figure 6, GA has rich pore structure, and its skeleton is wrinkled graphene. The pore structure of GA/AN nanocomposites is obviously less, and most pores are completely covered by AN filling, which makes it increase by 280.88 J/g on the basis of mechanical mixture, and the increase range reaches 50.8%. The reasons are as follows: First, graphene itself has a catalytic effect on the thermal decomposition of KP; Second, graphene is a special two-dimensional structure, which can effectively improve the dispersion of nano-catalysts, prevent agglomeration, and facilitate the adsorption and reaction between products and catalytically active sites during KP thermal decomposition; Third, graphene has excellent conductivity, and nano-oxides are loaded on graphene, which is equivalent to enhancing its conductivity and accelerating the electron transfer process; Fourth, graphene can be used as a thermal conductive medium in the thermal decomposition of KP because of its good thermal conductivity.

In conclusion, the application of nano-materials can significantly improve the decomposition thermal of pyrotechnics. Under the same material conditions, different mixing and preparation methods would also affect the decomposition heat to varying degrees, while the addition of carbon nano-materials can obviously improve the agglomeration phenomenon of nano-materials, make each component distribute evenly, and then greatly improve the decomposition heat of pyrotechnics, as shown in Table 2.

| Nanomaterial Classification | Nanoparticles | Particle Size/nm | Thermal Decomposition/ (kJ/g) | Increase Range | Researchers |
|-----------------------------|---------------|-------------------|-------------------------------|----------------|-------------|
| Metal Nanoparticles         | Cu            | 20 nm±            | 0.76                          | 172.7%         | Tan, et al. [10] |
| Metal Oxides and Their      | Fe$_2$O$_3$   | 8-25 nm±          | 5.095                         | 30.5%          | Ma, et al. [9] |
| Composite                   | FeO/Al        | 20 nm±            | 1.648                         | 95.7%          | Wang, et al. [24] |
| Nanoparticles               | TiO$_2$/APC   | 10 nm±            | 0.089                         | 18%            | Sherif, et al. [25] |
| Carbon-based Nanomaterials  | CuO/Al        | 100 nm±           | 1.52                          | 52.15%         | Wu, et al. [26] |
| Ga/AN                       | RGO/Fe$_3$O$_4$ | 30 nm±          | 0.533                         | 83%            | Sherif, et al. [25] |
| Nanomaterials               | GA/AN         | 70 nm±            | 50.8%                         | 100%           | Lan, et al. [27] |
| CoO/GA                      | 80 nm±        | 0.66              | 200%                          |                | Liu, et al. [28] |

4. Conclusion
In summary, the application of new nano-materials in pyrotechnics has made great progress, but there are also some problems in the development process, such as uneven dispersion of nano-particles in pyrotechnics, easy agglomeration of nano-particles and carriers, inefficient control of binding degree, and inaccurate quantification of catalytic effect of nano-catalysts. Therefore, the future application of nano-materials in pyrotechnics can be studied from the following directions:

a. Research and innovate the preparation methods and means of nano-materials, and reduce the particle size of nano-materials. At present, the particle size of nano-materials is mostly concentrated in tens to tens of nanometers, which is far from the 0.1 nm level of short-period metal atoms.
b. Innovate and explore the preparation methods of nano-agents and the technical means of combining with nano-carriers, so as to minimize the agglomeration of nano-particles, improve the degree of combination with carriers and make them uniformly distributed in the system.

c. Deepen the research and data analysis of catalytic mechanism of nanoparticles, and explore the lifting function of nanoparticles on reaction rate and effect, so as to achieve accurate and controllable catalytic effect.

d. Looking for green and environmentally friendly alternative nano-materials to reduce the pollution degree after pyrotechnic reaction. In addition, if the manufacturing cost of nano-materials can be reduced and mass production can be realized in the future, it will play an extremely important role in promoting the miniaturization and precision of pyrotechnic products.

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