

Surface Modification of AlN Powders By N-Vinylpyrrolidone Copolymers For Anti-Hydrolysis

Jianjun Xie (✉ xiejianjun@shu.edu.cn)

Shanghai University

Yu Wang

Shanghai University

Shun Wang

Shanghai University

Guangcheng Yu

Shanghai University

Tun Wang

Shanghai University

Lei Zhang

Shanghai University

Ying Shi

Shanghai University

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Abstract

By simply reacting raw aluminum nitride (AlN) powders with water-soluble N-vinylpyrrolidone (NVP) copolymers in absolute ethanol at a certain temperature to prepared hydrolysis-resistant AlN powders. The surface chemical structure and phase composition of the AlN powder has been investigated by using Fourier Transform Infrared Spectroscopy (FTIR) and X-ray diffraction (XRD), the surface morphology and element distribution of AlN were observed by the scanning electron microscopy (SEM) and X-ray energy dispersive spectroscopy (EDS), and the pH change of modified AlN powders was measured at different temperatures by soaked in deionized water to directly characterize the hydrolysis resistance. It was found that the surface of AlN was successfully coated and wrapped by seven kinds of N-vinylpyrrolidone polymers and effectively improved the hydrolysis resistance to varying degrees, among which N-vinylpyrrolidone-maleic anhydride copolymer and N-vinylpyrrolidone-itaconic acid copolymer have the best anti-hydrolysis effect which can maintain the properties of AlN unchanged after being soaked in water at 25°C for 24 hours.

1 Introduction

Aluminum nitride (AlN) ceramics display a series of excellent physical properties such as high thermal conductivity (100-320W/m·K), very low coefficient of thermal expansion ($4 \times 10^{-6}/K$) and dielectric constant (8.8 (1MHz)), which can be applied to substrate material for high-power device packaging and heat dissipation [1–7]. Meanwhile, the mechanical strength (350Mpa) of AlN ceramics is higher than Al_2O_3 and BeO ceramics, and it also has good thermal stability and corrosion resistance which make it to be one of the most promising materials in refractories, structural materials and surface protection of crucibles [8–12]. In addition, high purity AlN ceramics are transparent and can be used as optical devices [13]. However, AlN powders could easily undergo a hydrolysis reaction with water and lead the formation of $AlOOH$ or $Al(OH)_3$, which affects subsequent ceramic sintering and causes performance degradation [14–17]. Moreover, it can only apply non-water-based molding process that the organic solvent used in the molding process is not only expensive, but also has problems of environmental pollution [18, 19].

The hydrolysis resistance of AlN powders can effectively be improved by surface modification, including heat treatment, acid modification, surfactant modification, coupling agent modification, etc. [20–25]. Among K Krnel et al used phosphoric acid to modify AlN and proposed that a layer of phosphoric acid molecular layer was formed by esterification reaction of hydroxyl groups on the surface of AlN, which could effectively wrap AlN to prevent hydrolysis within 72h [26]. However, the phosphorus (P) element is not easy to remove in the subsequent sintering process of AlN ceramics and adversely affects the properties of the ceramics. It is necessary to further explore the other ways to resist hydrolysis of AlN powders.

Polyvinylpyrrolidone (PVP) is a water-soluble polymer which has good dispersion in water, non-toxic and non-irritating [27, 28]. Its monomer N-vinylpyrrolidone (NVP) can combine with other monomers which have different functional groups for achieving different functions. When the NVP copolymer contains

grafted functional groups such as acid anhydrides, carboxyl groups and amino groups, it can undergo esterification reaction with the active site hydroxyl groups on the surface of the AlN powders and lead the polymer chain wraps around the AlN powders. Meanwhile, the extension of the NVP hydrophilic group relies on steric hindrance effect in water to achieve the hydrolysis resistance of AlN powders. And the NVP copolymer generates CO₂ and other gases to volatilize in the subsequent sintering process of AlN ceramics at high temperatures, which does not affect the properties of ceramic.

In this study, seven kinds of water-soluble NVP copolymers with different functional groups were reacted with the hydroxyl groups of AlN powder to form a coating layer, and the pH changes of the seven kinds of modified AlN aqueous solutions were monitored at different temperatures to evaluate the effect of inhibiting hydrolysis. Among them, N-vinylpyrrolidone-Maleic anhydride copolymer (NVP-MAH) and N-vinylpyrrolidone-Itaconic acid copolymer (NVP-IA) modified AlN had the best anti hydrolysis effect at different temperatures. FT-IR, XRD, SEM and EDS were used to characterize the surface of the modified AlN chemical structure and morphology.

2 Experimental Methods

2.1 Raw Materials

AlN powders (Grade H, Tokuyama, Japan, average particle size = 0.5 μm, w(N) = 27.51%) were used in this study. N-vinylpyrrolidone copolymer such as N-vinylpyrrolidone-Maleic anhydride copolymer (NVP-MAH), N-vinylpyrrolidone-Itaconic acid copolymer (NVP-IA), N-vinylpyrrolidone-Itaconic acid-Lauryl methacrylate copolymer (NVP-IA-LMA), N-vinylpyrrolidone-Hydroxyethyl methacrylate copolymer (NVP-HEMA), N-vinylpyrrolidone-Acrylamide copolymer (NVP-AM), N-vinylpyrrolidone-Hydroxymethyl acrylamide copolymer (NVP-HAM) and N-vinylpyrrolidone-Itaconic acid-Methacrylic acid polyethylene glycol single methyl ether ester copolymer (NVP-IA-MAAMPEG) (Yoking Corp., China) were used as coating agents to protect the AlN powders. Absolute ethanol (mass fraction ≥ 99.5, chemically pure) was used as solvent.

2.2 Sample Preparation

As shown in Fig. 1, 10g AlN powders was dissolved in 20ml anhydrous ethanol

and then dispersed by magnetic stirring at 80°C for 30 min. 1g of NVP copolymer was dissolved in 20ml absolute ethanol and stirred at 60°C for a period of time to form a translucent solution. The uniformly dispersed NVP copolymer solution was slowly added to AlN solution and then stirred at 80°C for 4 h to obtain the white suspension product. The white precipitate was obtained by ethanol dispersion, washing and centrifugation, and then dried in a drying oven for 4 h at 80°C to obtain surface modified AlN powders.

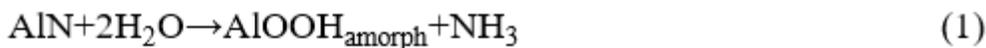
2.3 Performance testing

The hydrolysis test was carried out in a suspension of deionized water containing 2wt% AlN powder. In these tests, 2g of modified AlN powder was dissolved in 100ml deionized water and stirred to form suspension solution. Then, it was placed in the oven at 25°C, 40°C, 60°C and 80°C respectively to measure the relationship between pH (SevenCompact, Mettler Toledo Co.) value change and time. The surface chemical structure of AlN was determined by Fourier Transform Infrared Spectroscopy (Thermo Scientific Nicolet iS 50, ATR). The the phase change of AlN was measured by X-ray diffraction (D/MAX2200, 3KW). The surface morphology of AlN particles was measured by scanning electron microscope (EM-30, COXEM, South Korea), and the element content was measured by Energy-dispersive X-ray spectroscopy (ULTim Max Compact, oxford, UK).

3 Results And Discussion

3.1 Characteristics of the treated AlN in aqueous media

AlN powders have high reaction activity with water, and the hydrolysis mechanism follows the following equation [29]:



Firstly, initial AlN powders was found to be react with water to form amorphous phase $\text{AlOOH}_{\text{amorph}}$ and NH_3 , then $\text{AlOOH}_{\text{amorph}}$ reacts with water under certain temperature to form crystalline $\text{Al}(\text{OH})_3$, and NH_3 reacts with water to produce OH^- which leads to the increase of pH value of suspension liquid [30]. So the degree of hydrolysis can be judged by characterizing the change of pH value.

As shown in Fig. 2, the pH changes with time of as-received AlN powders and different NVP copolymers modified AlN powders in water at different temperature. As shown in Fig. 2(a), the untreated AlN powders were hydrolyzed continuously in the first 5 hours and the pH steadily increased to 10 at 25 °C, but the initial value of the other seven modified AlN powders remained unchanged. Within 5 to 12 hours, the pH of AlN powders which modified by NVP-MAH, NVP-IA, NVP-IA-LMA and NVP-IA-MAAMPEG remained unchanged, while the pH of NVP-HEMA, NVP-AM and NVP-HAM modified powders increased to about 8.5 and began to hydrolyze gradually. Among them, the pH of NVP-MAH and NVP-IA modified powders remained below 7 after 24 hours. This meant the anti-hydrolysis effect of the two modified AlN powders was the best among the seven NVP copolymers.

Water is more likely to react with AlN when the activation rate of water molecules increases with raising temperature, and the rate of hydrolysis of AlN powders is accelerated. In order to explore the anti-

hydrolysis effect of modified AlN powders at different temperatures, the pH changes of modified AlN powders were measured at 25 °C, 40 °C, 60 °C and 80 °C. The results show that the NVP-MAH and NVP-IA respectively modified AlN have best anti-hydrolysis effect regardless of any temperatures. Among them, maleic anhydride reacts in ethanol for a period of time to produce carboxylic acid groups after alcoholysis, which is easy to react with hydroxyl groups on the surface of AlN powders to form a coating layer for anti-hydrolysis.

As shown in Fig. 3(b), Fig. 3(c) and Fig. 3(f), the modification effect is poor in NVP-HEMA, NVP-AM and NVP-HAM polymers which structural monomers do not contain carboxylic acid groups. Although these polymers have some functional groups such as amino, imino and hydroxyl which are difficult to react with the hydroxyl on the surface of AlN powders, so that they cannot form an effective anti-hydrolysis coating.

In Fig. 3(d), Fig. 3(e), and Fig. 3(g), although all of these three types of itaconic acid copolymers contain carboxylic acid groups, their specific structures are different which lead to various modification effect. The steric hindrance effect of reaction groups raises with the increase of monomer types in polymer which hinders the dehydration condensation reaction between itaconic acid monomer and the surface hydroxyl group of AlN powders, so that AlN powders are not easy to be wrapped by polymer chains to form an effective anti-hydrolysis coating. Thus the anti-hydrolysis effect of modified AlN powders decreases with the increase of monomer types, which is testified by our experimental results: NVP-IA>NVP-IA-LMA>NVP-IA-MAAMPEG (anti-hydrolysis effect).

3.2 Structure composition and morphology characterization

NVP-MAH and NVP-IA with the best anti hydrolysis performance were selected for characterization from seven kinds of NVP copolymer. In Fig. 4 and Fig. 5, the surface functional groups of these two kinds of NVP-MAH and NVP-IA modified AlN powders were characterized by Fourier transform infrared spectra, and the region where the main characteristic functional groups exist is enlarged from 1900-1000 cm^{-1} as shown in Fig. 4(b) and Fig. 5(b). It has been reported that 600-900 cm^{-1} is a strong absorption peak of Al-N, and there exists simultaneously a weak absorption peak of Al-N at 1339 cm^{-1} [27, 31, 32]. In Fig. 4(a), there are two stretching characteristic peaks of anhydride in NVP-MAH at 1850 cm^{-1} and 1779 cm^{-1} , the characteristic peak of vibration of C=O from VP can be observed at 1664 cm^{-1} and 1287 cm^{-1} is the stretching vibration absorption peak of C-N. Since the acid anhydride in NVP-MAH reacts with the hydroxyl groups on the surface of aluminum nitride to form ester bonds, it can be observed that the modified AlN has a characteristic peak of 1718 cm^{-1} aliphatic ester bonds replacing 1850 cm^{-1} and 1779 cm^{-1} anhydride in Fig. 4(b). At the same time, the C=O tensile vibration absorption peak and the flexural vibration characteristic peak of C-H in NVP-MAH also respectively appeared at 1647 cm^{-1} and 1394 cm^{-1} , which proved that the surface of AlN was wrapped with a layer of NVP-MAH polymer. In Fig. 5(a), it can be found that 1723 cm^{-1} is the stretching vibration absorption peak of aliphatic carboxylic acid come from IA monomer, 1094 cm^{-1} and 1047 cm^{-1} are respectively C=O stretching vibration peaks in carboxylic

acid, and 1655 cm^{-1} is C=O characteristic peak of stretching vibration of VP monomer. After the NVP-IA modified AlN powder is hydrolyzed at 25°C for 24 hours, it can be observed from Fig. 5(b) that the carboxylic acid C=O stretching vibration absorption peak of 1721 cm^{-1} and the C-H bending vibration absorption peak at 1394 cm^{-1} are still exists, which indicates that NVP-IA is successfully coated on the surface of AlN powders.

The X-ray diffraction pattern in Fig. 6 presents the crystallinity between the pure AlN and treated powders. It can be observed that the untreated AlN have no same diffraction peak like the AlN standard card (PDF #25-1133), which is completely hydrolyzed after soaked in water for 24 hours. Meanwhile, it is found that NVP-MAH or NVP-IA treated AlN powders which were reveals the same major diffraction peaks as pure AlN after soaked in water for 24 hours. These results indicate that surface modification of AlN which coating with polymer doesn't affect the crystallinity of AlN, and these AlN powders modified by NVP-MAH and NVP-IA respectively have good hydrolysis resistance.

In Fig. 7(a), the as-received AlN powders have smooth surface and uniform particle size with a diameter of about 800 nm. After hydrolysis for 4h, the spherical size of the as-received AlN powders is aggregated to form large rod-like structure with a length of about 10 μm as shown in Fig. 7(b). As shown in Fig. 7(c) and Fig. 7(e), the AlN powders coated with NVP-MAH and NVP-IA basically remained spherical, but the polymer chains were easily intertwined with each other which cause the powders were easily bonded together to form aggregation. After these treated AlN powders are soaked in deionized water at 25°C for 24 hours, the surface morphology of the treated powder is still smooth spherical particles without rod or flake formed as shown in Fig. 7(d) and Fig. 7(f), which indicate that the two modified AlN powders are not hydrolyzed and have excellent hydrolysis resistance.

Fig. 8 and Fig. 9 show the X-ray energy spectrum and the results of the element distribution analysis of NVP-IA and NVP-MAH modified AlN powders after immersing in water at 25°C for 24 hours. In Fig. 8(d), Fig. 8(e), Fig. 9(d) and Fig. 9(e), it can be observed that the C and O elements contained in the treated AlN powders are evenly distributed on the surface of AlN powders, which means there is no element aggregation phenomenon, indicating that the NVP copolymer is uniform on the surface of AlN package. Due to the treated AlN powders are sprayed on the aluminum foil for EDS test, so the content of Al is relatively higher than N element as shown in Fig. 8(f) and Fig. 9(f), but the total atomic percentage of aluminum and nitrogen is about one to one. In table 1 and table 2, the content of oxygen element is very low which basically comes from NVP copolymers, it shows that the modified AlN powders does not form AlOOH or $\text{Al}(\text{OH})_3$ to lead a significant increase in oxygen element after soaking in water for 24 hours, it has been verified that the modified AlN has good hydrolysis resistance.

Table 1 Percentage of elements in NVP-IA modified AlN powders

Elements	Weight percentage	Wt%Sigma	Atomic percentage
Al	68.30	0.35	52.55
N	25.65	0.26	38.01
C	4.13	0.36	7.13
O	1.92	0.14	2.30
Total	100.00	1.00	100.00

Table 2 Percentage of elements in NVP-MAH modified AIN powders

Elements	Weight percentage	Wt%Sigma	Atomic percentage
Al	64.97	0.44	48.71
N	29.03	0.34	41.92
C	4.26	0.46	7.18
O	1.74	0.15	2.20
Total	100.00	1.00	100.00

4 Conclusion

AIN powders modified by seven kinds of NVP copolymers can resist hydrolysis to varying degrees at different temperatures, but the resistance to hydrolysis decreases with the increase of temperature. Among them, NVP-HEMA, NVP-AM and NVP-HAM are difficult to form an anti-hydrolysis coating due to the lack of hydroxyl groups. In addition, the anti-hydrolysis effect is testified by our experimental result that NVP-IA>NVP-IA-LMA>NVP-IA-MAAMPEG, because the steric hindrance effect also increases with the raise of monomer types which lead the hydrolysis resistance effect decreases. Among the seven polymers, NVP-MAH and NVP-IA have the best anti-hydrolysis effect as a result of carboxylic acid groups can undergo esterification with hydroxyl groups on the surface of AIN powders. These two treated powders can maintain the properties of AIN without being hydrolyzed after soaking in water at 25°C for 24 hours.

Declarations

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Figures

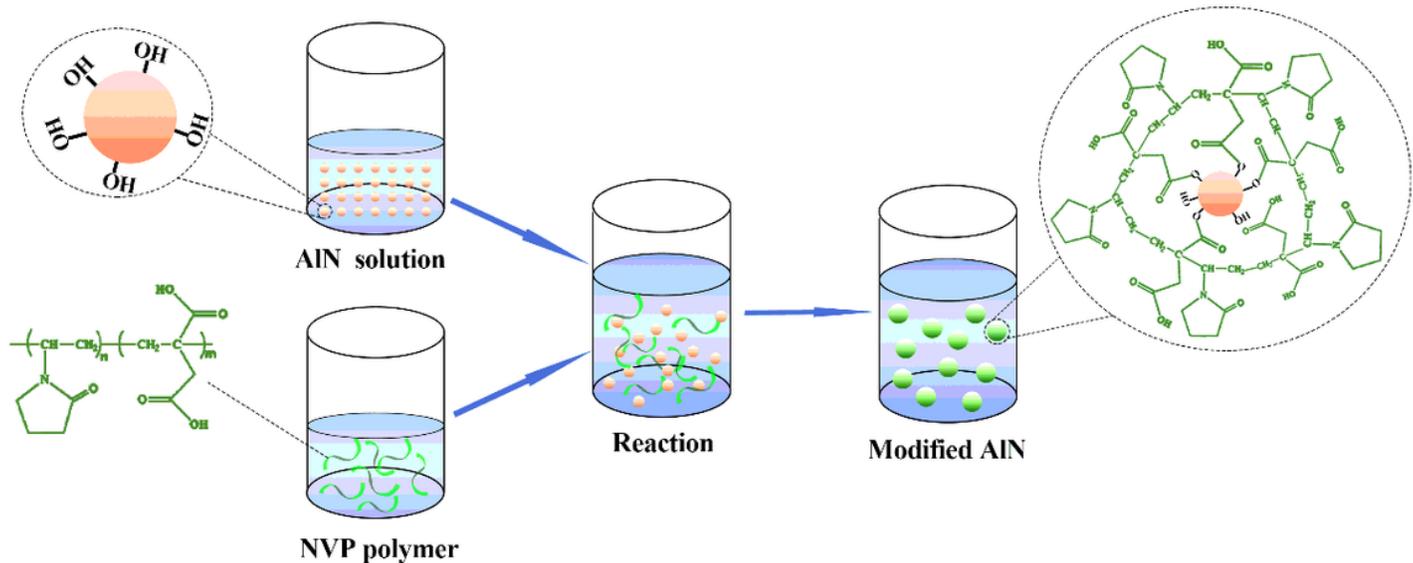


Figure 1

Schematic illustration of preparation of NVP polymers modified AIN powders

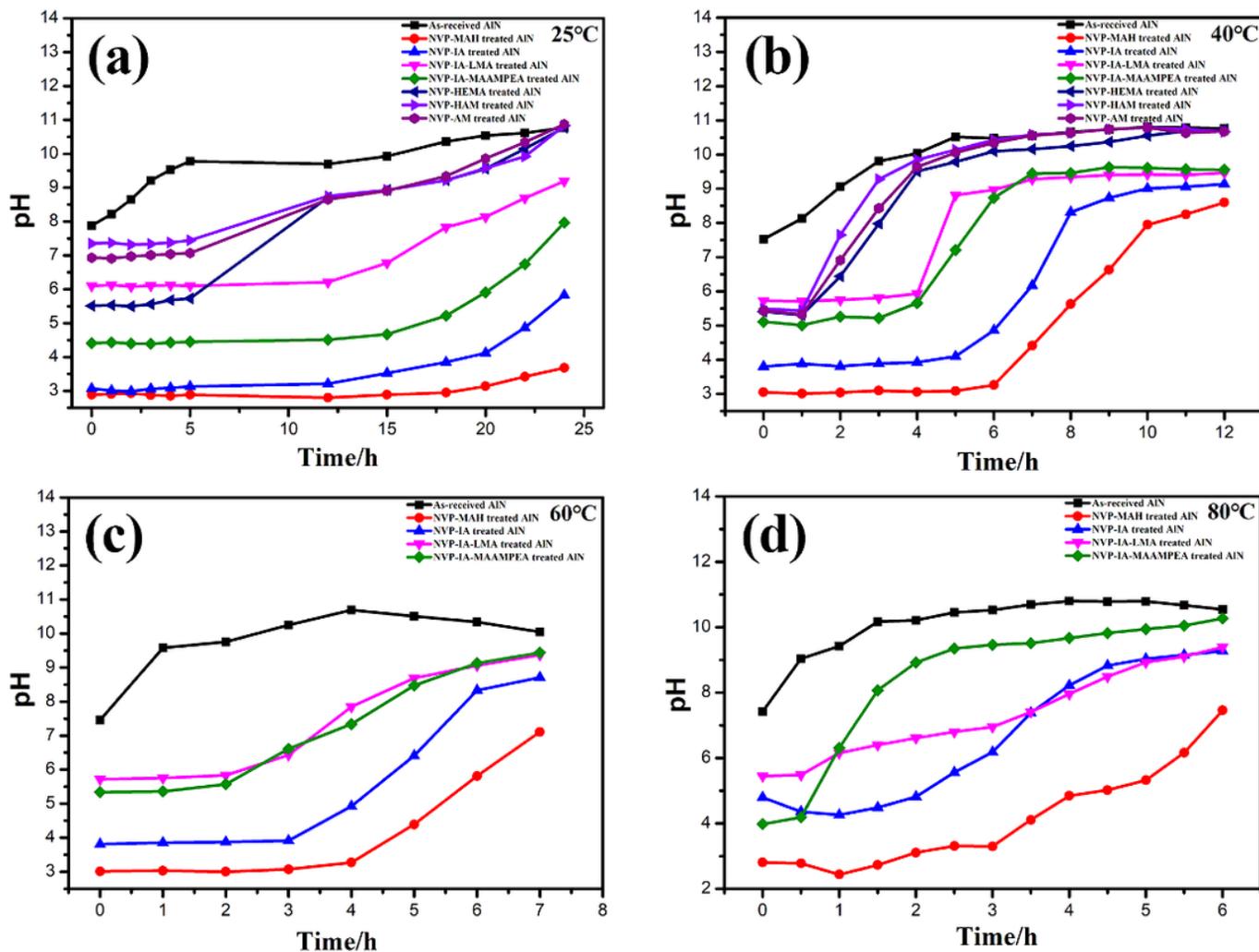


Figure 2

pH value-time curve of as-received AIN powders / modified AIN powders at different temperature (a) pH value-time curve of as-received AIN powders / seven modified AIN powders at 25 °C (b) pH value-time curve of as-received AIN powders / seven modified AIN powders at 40 °C (c) pH value-time curve of as-received AIN powders / four modified AIN powders at 60 °C (d) pH value-time curve of as-received AIN powders / four modified AIN powders at 80 °C

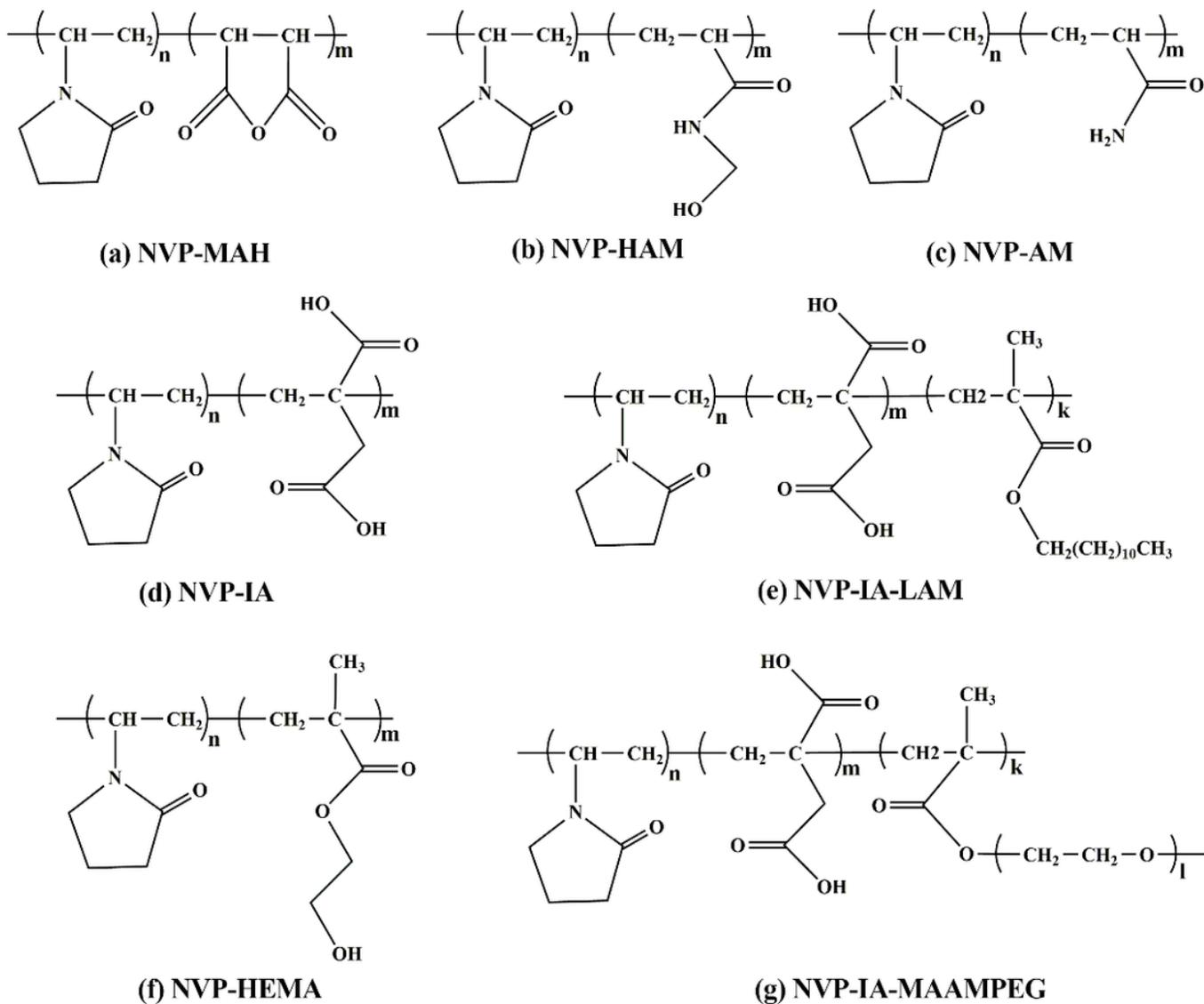


Figure 3

Molecular structures of seven NVP polymers

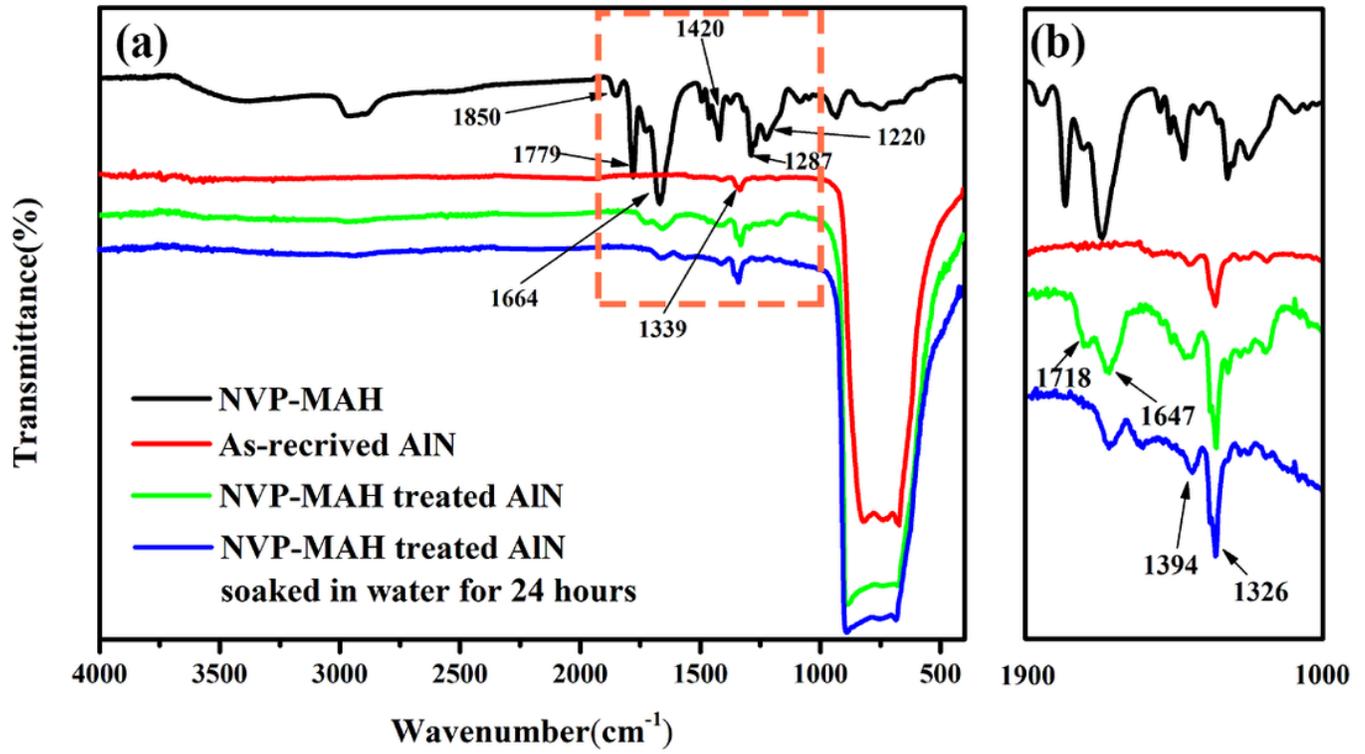


Figure 4

FT-IR spectra of as-received AlN powders / NVP-MAH modified AlN powders

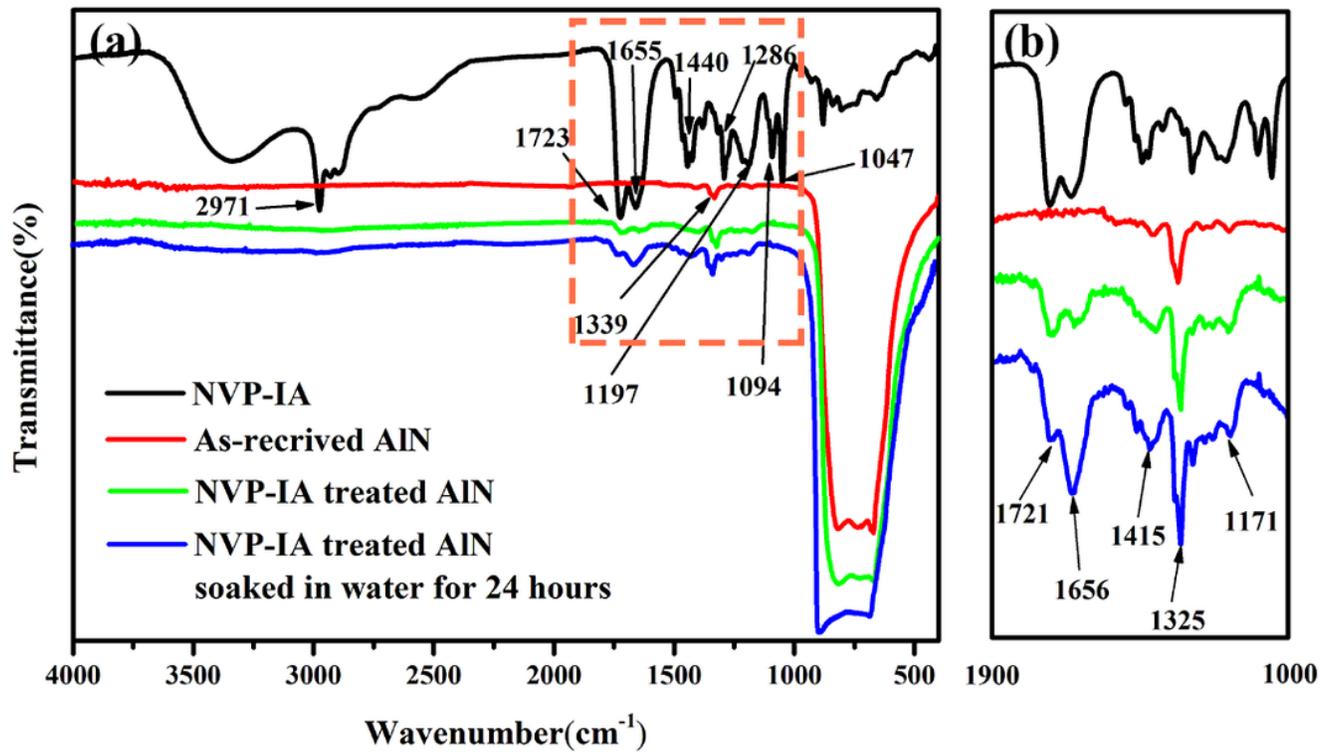


Figure 5

FT-IR spectra of as-received AlN powders / NVP-IA modified AlN powders

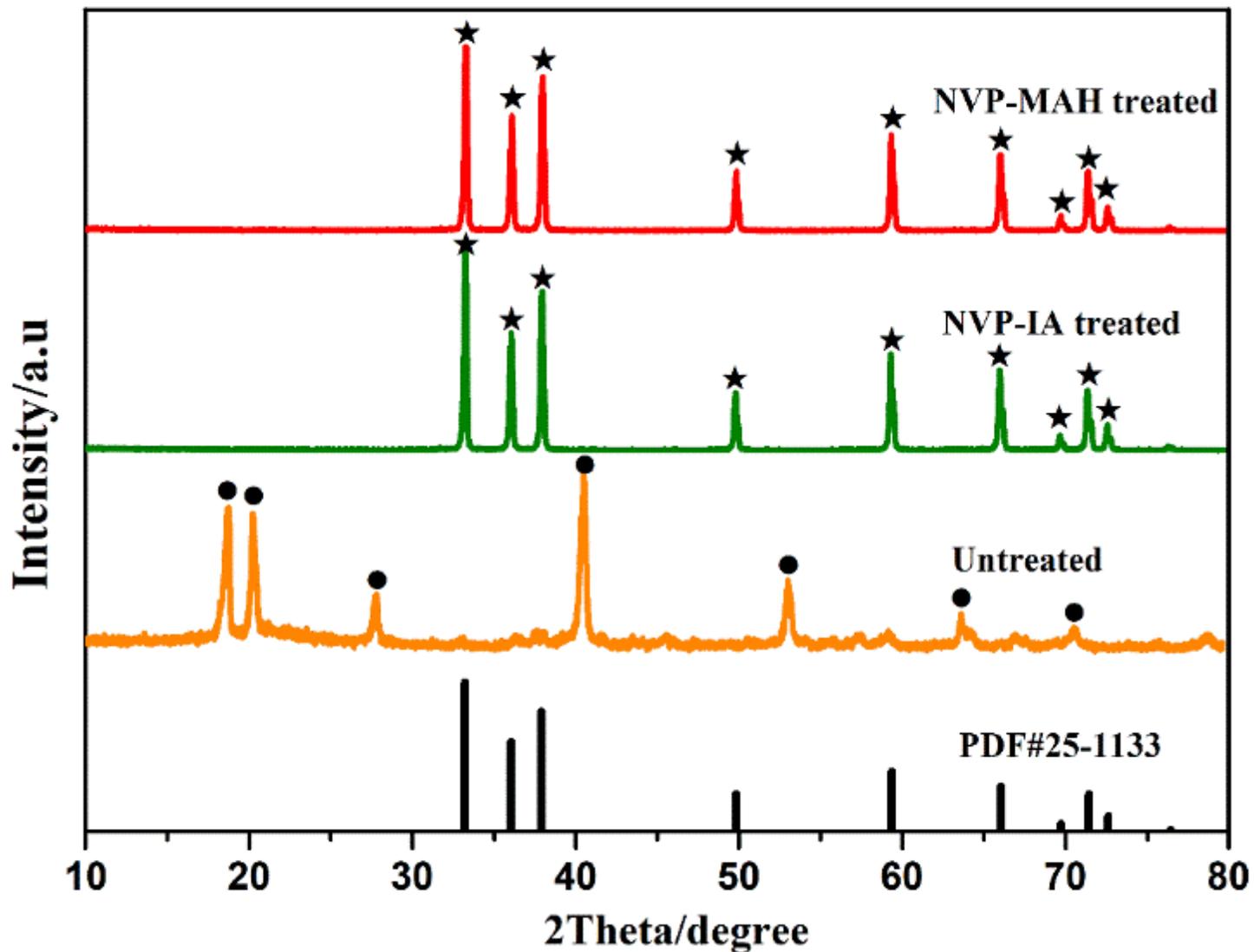


Figure 6

XRD pattern of as-received AlN powders / modified AlN powders after soaking in water at 25 °C for 24 h

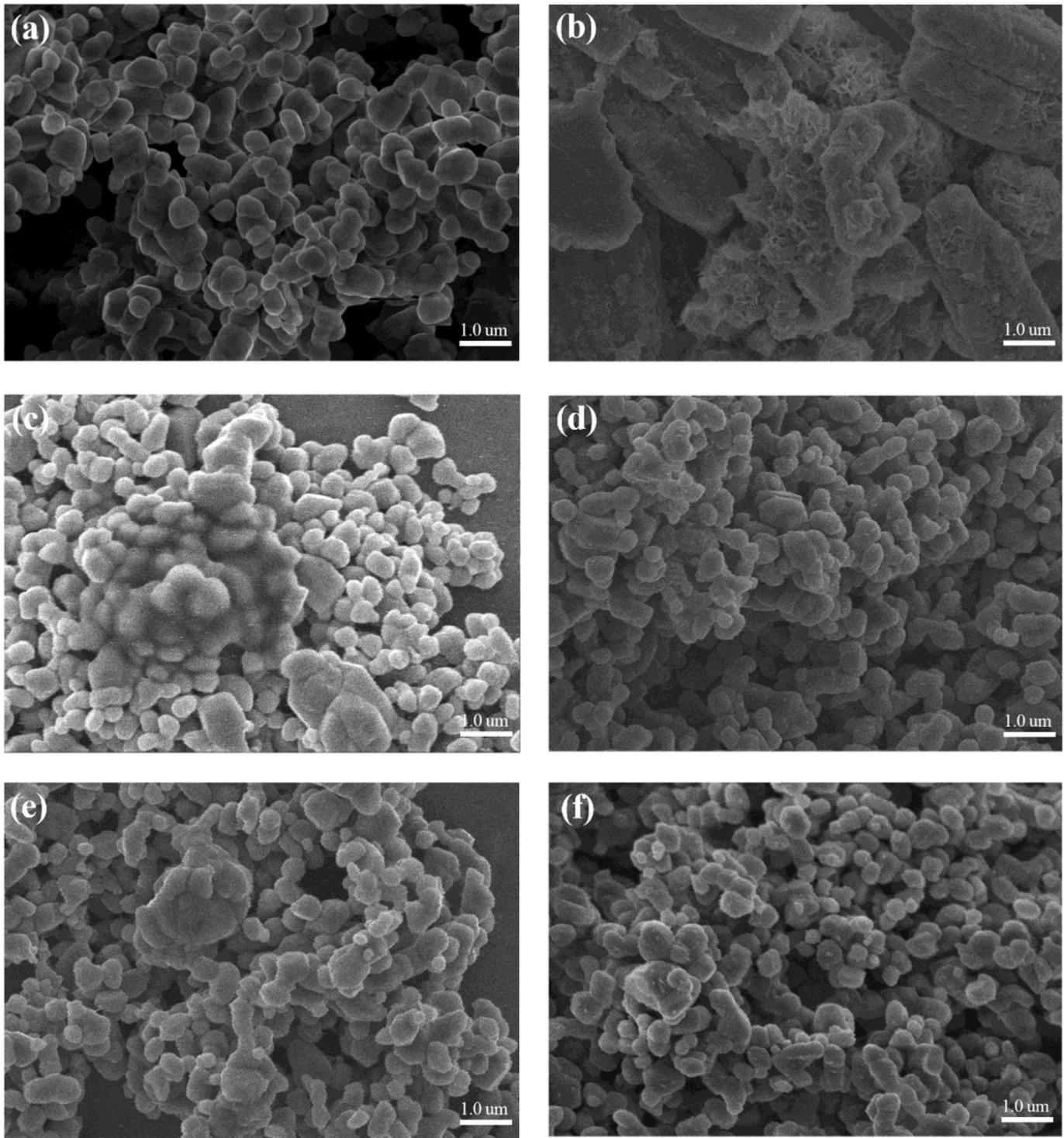


Figure 7

SEM micrographs of as-received / modified AlN powders (a) SEM micrograph of as-received AlN powders (b) SEM micrograph of as-received AlN powders after soaking in water for 4 hours at 25°C (c) SEM micrograph of AlN powders modified by NVP-MAH polymers (d) SEM micrograph of AlN powders modified by NVP-MAH after soaking in water for 24 hours at 25°C (e) SEM micrograph of AlN powders

modified by NVP-IA polymers (f) SEM micrograph of AlN powders modified by NVP-IA after soaking in water for 24 hours at 25°C

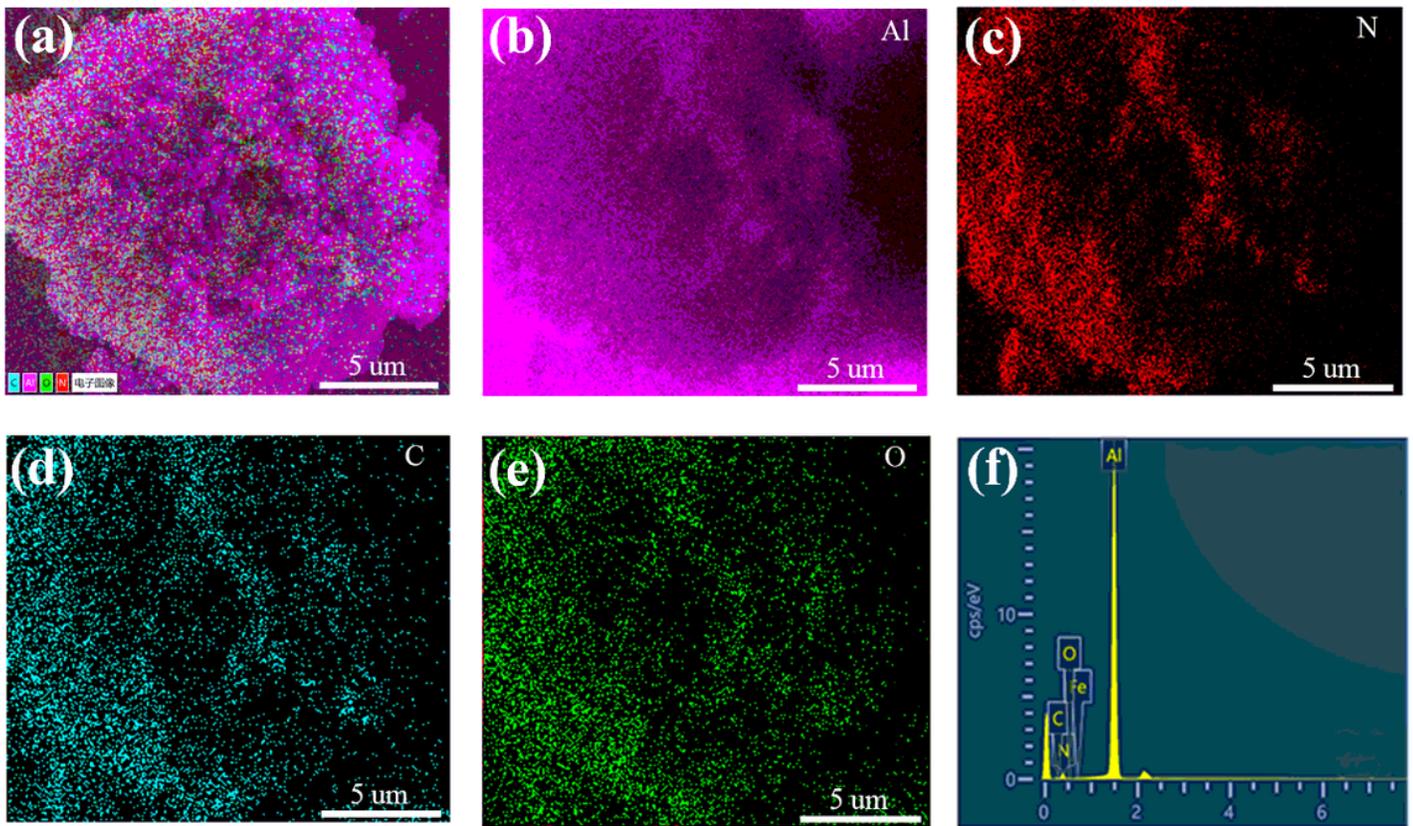


Figure 8

EDS graphs of AlN powders modified by NVP-IA after soaking in water for 4 hours at 25°C (a) EDS layered-graph of AlN powders modified by NVP-IA; (b) EDS graph of Al distribution; (c) EDS graph of N distribution; (d) EDS graph of C distribution; (e) EDS graph of O distribution; (f) Elements distribution of NVP-IA modified AlN powders.

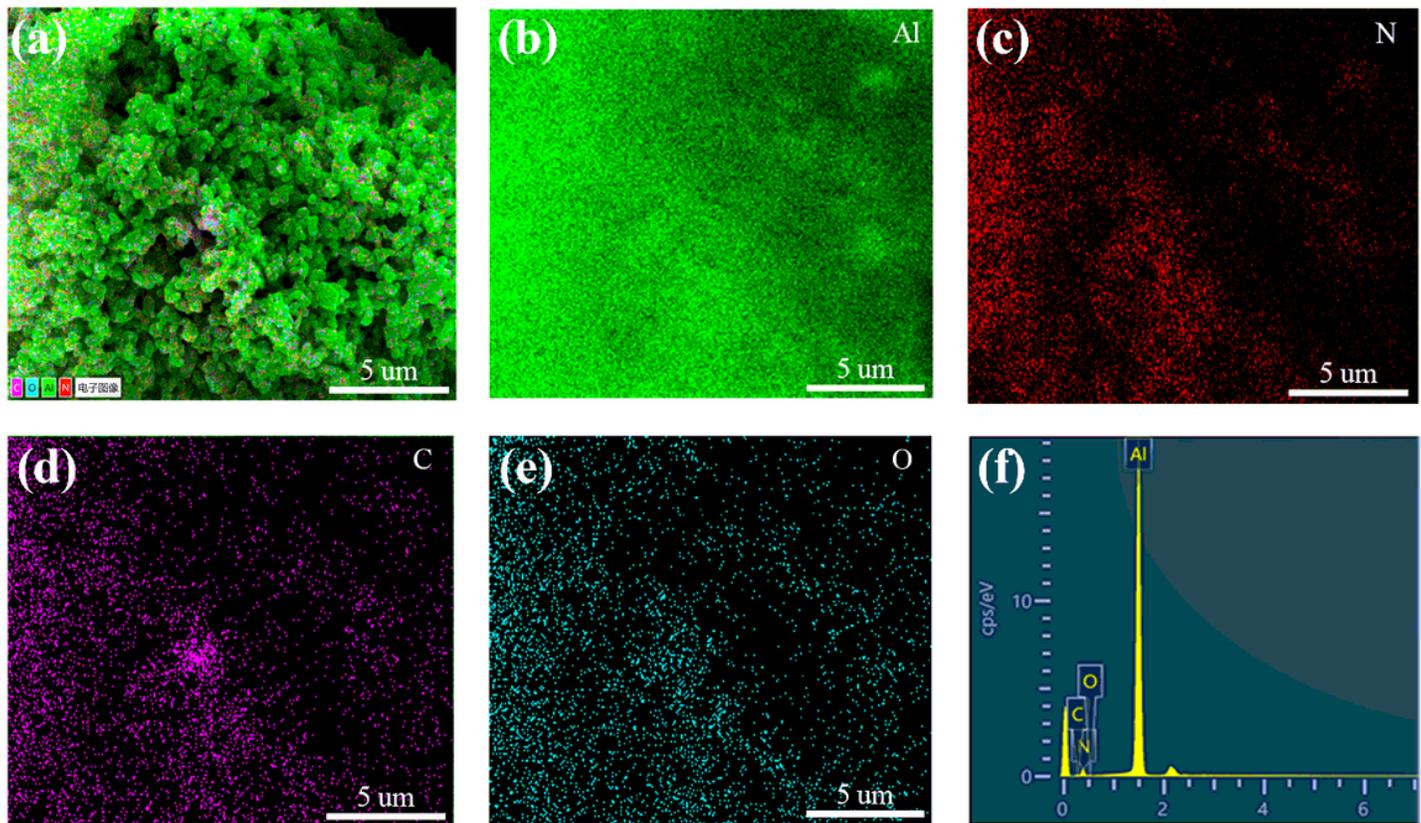


Figure 9

EDS graphs of AlN powders modified by NVP-MAH after soaking in water for 4 hours at 25°C (a) EDS layered-graph of AlN powders modified by NVP-MAH; (b) EDS graph of Al distribution; (c) EDS graph of N distribution; (d) EDS graph of C distribution; (e) EDS graph of O distribution; (f) Elements distribution of NVP-MAH modified AlN powders.