Review Article

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A mini-review of three-dimensional network topological structure nanocomposites: Preparation and mechanical properties

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Abstract: Three-dimensional (3D) network topological structure composite (3D-NTSC) is a new type of composite in which both the matrix phase and the reinforced phase have 3D continuous network structure and interweave with each other. Different from fiber reinforcement and particle reinforcement, the reinforcing phase in the composite material runs through the whole matrix phase in a continuous form without destroying the topological continuity of the matrix phase, so that each matrix phase in the composite material can not only maintain its own excellent performance, but also can cooperate with each other and complement each other, so that the composite material can play a greater performance advantage. In recent years, 3D-NTSC has attracted the attention of researchers and has been widely used in practical production. At present, there are no comprehensive articles summarizing the research progress of this kind of materials. In this review, we discussed the recent progress of the preparation methods, including natural flow method, vacuum infiltration process, pressure filling method, in situ filling method, and co-building method. Furthermore,

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research progress on mechanical properties and some regular results, shortcomings, challenges, and prospects of 3D-NTSC were also put forward, which would be helpful to people working in the related fields.

Keywords: three-dimensional network, topological structure, nanocomposite materials, preparation methods, mechanical properties

1 Introduction

With the progress of society and the development of science and technology, traditional homogeneous materials have been difficult to meet actual needs [1,2]. Therefore, how to prepare high-performance new composite materials has become a current hot topic. In recent years, Three-dimensional (3D) network topological structure composite (3D-NTSC) with low density, high specific modulus, high specific strength has been widely used in fields such as the preparation of structural loadbearing components [3], heat conduction [1], oil-water separation [4], electrical conduction [5], and energy storage due to its special reinforcement method and structures. 3D-NTSC is a composite material composed of two or more phases, and each phase is three-dimensionally connected through a microstructure, and the characteristics of each component in the network structure remain basically unchanged. If one phase is removed, the remaining phases will still maintain structural integrity and loadbearing capacity [6,7]. 3D-NTSC combines the advantages of three-dimensional braided materials and ordinary composite materials, especially when subjected to external forces, the network structures are mutually restrained and the interaction force is strengthened, and the two phases can be combined well, which is not achieved by many traditional materials. It is this special three-dimensional network topology that makes composite materials have isotropic properties and exhibit excellent physical

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and mechanical properties, which are often superior to traditional fiber or particle reinforced composite materials, providing new options for applications in many fields [8–11].

In practice, the structural mechanics of 3D-NTSC play a very important role, which determines whether it can be used in engineering. However, the complete theory has been developed to explain and predict the strength and structure of final composite materials, it is still a big challenge to effectively combine theory with practice. Although great achievements have been made in the research of 3D-NTSC, there is no instructive review of the research progress. Therefore, this article will briefly review the preparation methods and mechanical properties of 3D-NTSC. Furthermore, some shortcomings, challenges, and prospects of 3D-NTSC are also put forward, which will be helpful to people working in the related fields.

2 Composite structure

Composite materials can be divided into the following three types according to the reinforcement method: fiber reinforcement [12], particle reinforcement [2], and three-dimensional network reinforcement [13]. Particle-reinforced composite materials are mainly based on carbide, nitride, graphite, and other particle-reinforced metals or alloys. Although the material has good isotropy, the processing and preparation process are relatively simple, its characteristic is that the particles used for reinforcement are discontinuous, and the rigidity and strength are slightly lower than that of continuous fiber-reinforced materials, so its application will be limited. While the fiber-reinforced composite material is composed of various

fiber reinforcements placed in the matrix material, which can provide good hardness, strength, and stability to the whole material.

Their internal structure is shown in Figure 1, from which it can be seen that the particles and fibers are uniformly dispersed in the matrix. When the composite material is damaged by external force, they can hinder the crack propagation in the form of bridging at the crack development, which can achieve the purpose of improving the mechanical properties of the material to a certain extent. However, the particles of the reinforcing phase do not contact each other, and there is no interaction, and a certain destructive strength can make the cracks develop rapidly. On the contrary, if the particles or fibers are connected to each other, when the composite material is damaged by external force, the stress can be transferred and buffered along the reinforcing fillers. It is conceivable that the enhancement effect brought by this enhancement method far exceeds that of the dispersed enhancement phase, which is the advantage of the three-dimensional network enhancement. In addition, since the size of the reinforcing phase of the composite material is generally required to be small enough, this also makes the reinforcing phase easy to agglomerate without being dispersed. The three-dimensional topology network reinforcement structure itself is a continuous phase, and there is no dispersion problem, which is also an advantage of the threedimensional network enhancement [14].

3 3D-NTSC preparation

Based on the special structure composition of 3D-NTSC and many advantages, how to achieve effective preparation

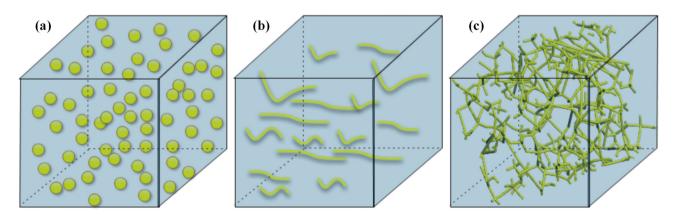


Figure 1: Schematic diagram of the internal structure of composite materials with different reinforcement methods: (a) particle reinforcement; (b) fiber reinforcement; and (c) three-dimensional network reinforcement.

becomes a key point. The construction method of the structure has a long origin, such as the mud wall built by the combination of straw and mud, reinforced cement composite structure. The prepared materials play an important role in load-bearing, electrical, and thermal conductivity. At present, the preparation methods of 3D-NTSC are mainly divided into two categories. One is the pre-forming matrix method, in which the matrix is prepared first and then the filling phase is introduced. The other is co-construction method, forming the matrix phase and reinforcement phase at the same time. The following is a brief summary and discussion of the related research on the preparation of 3D-NTSC according to the two methods.

3.1 Preformed matrix method

The preforming matrix method means that the matrix phase is preformed and then the filling phase is introduced in a specific way. Remarkably, the preformed matrix phase must present a three-dimensional network structure to give space for the filling phase. The preformed matrix phase can be prepared in a variety of ways, it can be obtained from the natural environment, such as natural sponge and loofah. In addition, it can also be prepared artificially, such as aluminum foam (AF), nickel foam, and aerogel. According to the introduction mode of reinforcement phase, it can be divided into four kinds, natural flow method, vacuum infiltration process [15], pressure filling method [16], and in situ filling method [17].

3.1.1 Natural flow method

The natural flow method is a simple molding method, usually by introducing the filling phase into the matrix phase in the natural state, without a vacuum or highpressure environment. Therefore, there are certain requirements on the filling phase, such as good fluidity and wettability [18-20]. The first reported preparation method of 3D-NTSC is the co-continuous ceramic composites developed by Breslin. He burned ceramics into threedimensional continuous skeleton structure as the matrix phase, and poured aluminum liquid as the filling phase in the sintering process to get the final composite [21]. This method has also been studied by other researchers. Aghajanian et al. made use of the fluidity of alumina to fill alumina naturally in the aluminum matrix [22], and Hong et al. filled the Cu matrix with 83 vol% of TiB₂ to produce a double-continuous TiB₂/Cu matrix composite with good thermal shock resistance [23]. In addition to the simple introduction of the filler phase by the inherent three-dimensional network structure of the matrix, the three-dimensional network morphology of the matrix phase is also further studied. By adjusting the porosity, pore size, and pore density of the threedimensional network structure, the filling phase and matrix phase can be better matched and combined, and finally the composite material with better performance can be obtained. Chen et al. [24] prepared porous C_f/B₄C-C preforms by reactive melt filtration method, studied the influence of the pore structure of C_f/B₄C-C preforms on the microstructure and mechanical properties of C_f/SiC-ZrC-ZrB₂ composites, and prepared three-dimensional C_f/SiC-ZrC-ZrB₂ composites.

3.1.2 Vacuum infiltration process

The vacuum infiltration process refers to the method of introducing the filling phase in a vacuum state. In this state, the fluid extension is not hindered by gas, which can avoid the problem of insufficient contact caused by gas adhesion on the surface of the matrix phase. Therefore, the mechanical properties of the composites prepared are more stable. This method is mainly used for filling matrix materials with compact network structure and of small size, such as aerogel [25] (Figure 2a) and high-density sponge [26] (Figure 2b).

The good dispersion and order structure of the threedimensional matrix skeleton are beneficial to the improvement of the properties of the final composites. How to ensure the effective combination between the filling phase and the matrix phase is the key. Ni et al. used graphene oxide (GO) aerogel as matrix skeleton to adsorb a layer of poly (amidoamine) dendrimer as reductant and cross-linking agent on the surface of the skeleton and then impregnated it in epoxy (EP). EP was poured into the aerogel under vacuum condition to obtain the composite material. Compared with the GO composites directly mixed with EP, the mechanical properties (tensile strength and compressive strength increased by 120.9 and 148.3%, respectively) and thermal stability (glass transition temperature increased by 19°C) of the composites with aerogel content of only 0.20 wt% were significantly improved [27] (Figure 2c-f). It is also significant to select suitable filling phases for specific types of skeletons. Liu and Gong used vacuum seepage method to fill EP and polyethylene (PE) into aluminum

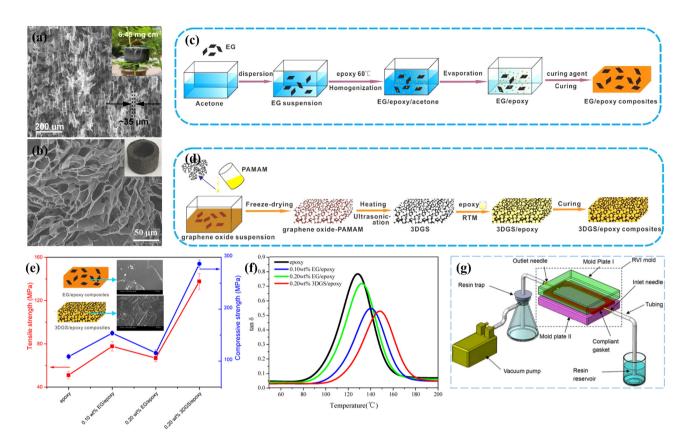


Figure 2: The SEM images and their corresponding objects of (a) aerogel [25] (Copyright 2021, American Chemical Society) and (b) high-density sponge [26] (Copyright 2018, Wiley). Preparation of EP/GO composites by (c) direct mixing and (d) vacuum seepage. (e) Mechanical properties and (f) thermal stability of Ep/graphene composites [27] (Copyright 2015, American Chemical Society). (g) Schematic diagram of high-speed seepage device [29] (Copyright 2016, Taylor & Francis).

foam, and compared the compression properties of the two [28]. The research on high-speed seepage device [29] (Figure 2g), to achieve excellent combination of matrix phase and reinforcement phase, and to prepare excellent comprehensive performance is the focus of subsequent research.

3.1.3 Pressure filling method

Different from the vacuum infiltration process, the pressure filling method needs to introduce the filling phase under the condition of applying external pressure, which requires the matrix skeleton network to have a certain strength. Therefore, the type of matrix material is limited when the composite material is prepared by this method. The matrix material is usually metal material, and the injection method is usually adopted to ensure the stable output of pressure.

During the pressure filling, the key to the successful preparation of the composite is to ensure the uniform force flow of the filling phase, maintain the original

matrix structure, and stabilize the forming of the composite. In order to ensure the realization of the above key points, designing the matching processing mode is of vital importance. Dukhan et al. took AF as the matrix to design an injection mold, as shown in the Figure 3a. AF was placed in the cavity of the mold, and the injection pressure was adjusted to make polypropylene (PP) fill AF evenly and completely. The final performance characterization shows that the structure of AF has a great influence on the properties of the composites, and the higher the porosity, the higher the bending modulus (Figure 3b) and bending (Figure 3c) strength of the composites [30]. Long et al. also used injection method to prepare a composite material with AF as the matrix and filled with polymers, and the bending and damping properties of the composite material were improved [31,32]. Su et al. also studied the mechanical and friction properties of the composite material prepared on this basis [33]. For the polymer material, on the basis of ensuring the performance requirements of the final composite, part of the embedded polymer in the matrix material may simplify the processing procedure to a certain extent. Therefore,

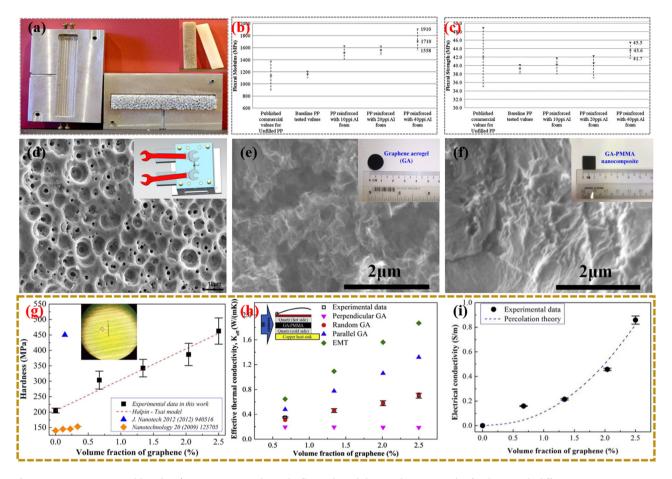


Figure 3: (a) Injection mold and AF/PP composite spline; (b) flexural modulus; and (c) strength of splines with different porosity [30] (Copyright 2010, Elsevier). (d) Schematic diagram of matrix and composite with micropore and nanopore structure morphology prepared by NMT technology [38] (Copyright 2021, Walter De Gruyter). (e) GA and (f) PMMA-filled GA digital photos and their FESEM pictures. The (g) hardness, (h) thermal conductivity, and (i) electrical conductivity of GA/PMMA composites were prepared with the variation in graphene content [40] (Copyright 2013, Multidisciplinary Digital Publishing Institute).

micro/nano forming technology (NMT) has also been widely used [34]. In this technology, the surface of metal substrate is treated by specific forming technologies, such as anodic oxidation, acid etching, and alkali etching, and micro/nano-holes with different sizes and morphology distribution are generated on the surface by controlling reaction conditions. Finally, the combination of metal substrate and resin is realized by means of pressure forming [35–37]. For example, Liu *et al.* implemented the anodic oxidation treatment strategy on the surface of the wrench to obtain a porous oxide film. By adjusting the voltage, temperature, and electrolyte concentration, micro- and nano-holes with specific diameter were obtained and a large hole sleeve keyhole structure was formed. Polyphenylene sulfide molecules are injected into the multiscale pores by the pinning effect, and the final composite material has good adhesion [38] (Figure 3d).

3.1.4 In situ filling method

In situ filling method is to construct a three-dimensional matrix network in advance and then synthesize the filling phase in the matrix. Compared with the previous methods, this method can also be applied to the matrix materials with denser network structure. This is because in a small space, it is difficult for macromolecular fluid or fluid with high viscosity to be fully infiltrated, so it is easier to achieve better filling by injecting small molecular monomers into the network structure and then polymerizing into polymer *in situ* to fill the network structure [39]. In this method, impregnation, decompression, and pressure are often used in operation.

Zeng *et al.* reported that after pre-polymerization of methyl methacrylate monomer to a certain viscosity, it was rapidly cooled to a temperature below 50°C to cease

the pre-polymerization, the graphene aerogel (GAs) are then pressed into the mixture and the mixture with the GAs immersed is kept at a low temperature to remove air bubbles (Figure 3e and f). Afterwards, the above mixture is maintained in a water bath at 50-55°C for 30 h until it completely solidifies. The obtained composites have a conductivity (Figure 3g) of 0.859 S/m, a microhardness (Figure 3h) of 462.5 MPa, and a thermal conductivity (Figure 3i) of 0.7 W/m K [40]. Aldosari et al. [41] also prepared polymethyl propene (PMMA/rGO) nanocomposites by in situ bulk polymerization using two different preparation techniques. According to the properties of the filling phase, the research direction of this method is to explore other types of filling phase (such as polydimethylsiloxane) and rapid filling method.

3.2 Co-building method

The preformed matrix method requires the filling phase to be filled into the matrix in the form of a fluid, which often requires heating or a longer curing and molding time for the filling phase, which makes the processing procedure more complicated and also increases the economic cost. The combination of the two phases during filling is affected by the processing conditions, and there may be non-contact interfaces or single-phase discontinuities, which have an unpredictable impact on the strength properties of the material. The co-building method is a method of forming the matrix phase and the filling at the same time, which greatly shortens the processing time, and can also make the filling phase and the matrix phase more uniform. The 3D-NTSC of porous foam composite prepared by co-building method exhibit excellent mechanical properties and thereby has a wide range of research [42,43]. For example, Cao et al. fabricated a novel nanofiber reinforced graphene aerogel (aPANF/GA), which has a 3D interconnected hierarchical microstructure with surface-treated PAN nanofiber as a support scaffold throughout the entire graphene network by ultrasonic dispersion and hydrothermal reaction, which is shown in Figure 4a. This 3D interconnected microporous aPANF/GA aerogel combines an excellent compressive stress and excellent conductivity, which can be proved by the increase in the intensity of the LED lamp as compression of the aPANF/GA increases. As shown in Figure 4b, the aPANF/GA introduced with aPAN nanofibers can withstand higher compressive stress than PAN/GA and GA, under the same compressive strain, which is proposed to be due to stronger connections between the alkaline treated PAN nanofibers and rGO nanosheets than untreated PAN nanofibers and rGO nanosheets [44]. In the other group, Wang et al. used a similar method to prepare boron nitride nanosheets (BNNS) and cellulose nanofibers (CNF) aerogel nanopapers [45]. BNNS/CNF aerogel nanopaper has excellent balanced thermal conductivity and electrical insulation performance, which was much higher than that of blended nanopaper, and holds great potential for a multitude of applications associated with electronics.

Besides aerogel, hydrogels of 3D-NTSC have a wide range of applications in the fields of bioengineering and electronic equipment due to their excellent mechanical robustness and conductivity. Si et al. presented a robust strategy for creating high water containing and superelastic NFHs with ordered cellular structures using sustainable alginate and electrospun nanofibers (Figure 4c) [46]. As shown in Figure 4d, the cellular architecture can be distilled to four levels of hierarchy at three individual dimension scales: unit cells, cell walls, and nanofibers. Furthermore, the mechanical properties of NFHs could be regulated simply by choosing different crosslinking cations (Figure 4e), and the Al-NFHs reached the mechanical strengths (16.04 kPa) for all samples. The inversion of the nanofibrous cells occurs isotopically upon compressive stress and suggests that this inversion is important for realizing near-zero Poisson's ratios, as illustrated in Figure 4f. Gels formed via metal-ligand coordination typically have very low branch functionality, as they consist of 2-3 polymer chains linked to single metal ions that serve as junctions. Thus, these materials are very soft and unable to withstand network defects such as dangling ends and loops. To solve the problems of gel formed by metal-ligand coordination, usually with very low branching functionality, Zhukhovitskiy et al. reported a new gel from polymer ligand and metal-organic cage (MOC) as a combination. They demonstrated that in polyMOCs with large junctions and a high number of loop defects it is possible to replace selectively defects with functional free ligands to imbue the material with a novel function without compromising mechanical integrity [47].

4 Research progress on mechanical properties of 3D-NTSC

As a composite material, its mechanical properties often become the criterion to consider whether it can be used in

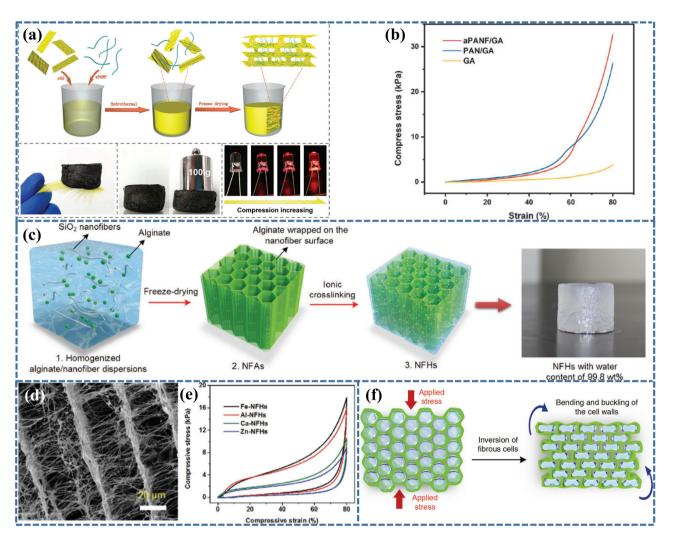


Figure 4: (a) The fabrication process, the mechanical property, and the conductivity of aPANF/GA. (b) Compressive σ - ε curve of GA, PAN/GA, and aPANF/GA 0.5–1 (ε form 0–80%) [44] (Copyright 2020, Wiley). (c) Schematic for creating high water containing and superelastic nanofibrous hydrogels (NFHs) with ordered cellular structures using sustainable alginate and electrospun nanofibers. (d) SEM image of NFHs demonstrating the hierarchical nanofibrous cellular architecture. (e) Compressive σ versus ε curves for NFHs cross-linked by various metallic cations. (f) Schematic demonstration of the inversion of the nanofibrous cell walls under compression [46] (Copyright 2017, Wiley).

practical application. Moreover, the advantage of 3D-NTSC is to learn from each other's strengths and make up for each other's weaknesses. Combining the advantages of each single-phase material, the material presents excellent performance. Its application is mainly focused on replacing the existing parts as structural components, so its mechanical properties are widely concerned. The mechanical properties of single-phase skeleton materials are often poor, but the mechanical properties can be improved after composite. Contrast with the particle and fiber reinforced way which developed maturely, the enhancement mechanism of this composite has no related reviews with guidance meaning, although there are some mechanics studies about compression and damping properties, bending [48], tensile [23] performances, but few can find all above in one report.

To represent the various types of mechanical studies of the composite, some studies are summarized in Table 1. It shows the excellent application prospect of 3D-NTSC.

Chen prepared AF composite materials filled with rosin, EP, and EP filled with nano-montmorillonite by natural infiltration method, and the results showed that their compression, damping, and impact resistance properties were improved after filling with resin [49]. In another group, the mechanical properties in bending tests of an integrated composite sandwich panel of AF and EP resin were studied through quasi-static four-point bending tests [50]. The mechanical properties of the integrated composite sandwich panel have been highly improved compared with those of the traditional sandwich panel. The specimens maintain good stability under bending, and no peeling-

Table 1: Mechanical properties of different 3D-NTSCs

Materials		Compression strength (MPa)			Flexural strength (MPa)			Tensile strength (MPa)			Ref.
Matrix	Filler	Matrix	Filler	Composite	Matrix	Filler	Composite	Matrix	Filler	Composite	
AF	Ер	5.8	66.2	42.7	_	_	_	_	_	_	[19]
AF	Еp	10.0	_	30.0	_	_	_	_	_	_	[31]
AF	PP	_	_	_	42.0	39.6	43.6	45.0	20.0	29.0	[28,48]
AF	SR^1	_	_	140.0	_	_	_	_	_	_	[52]
AF	POM	_	_	163.6	_	_	184.9	_	_	_	[30]
Ti ₃₅	Ep	50.0	80.0	120.0	_	_	_	_	_	_	[59]
GA	Еp	100.0	_	300.0	110.0	_	165.0	_	_	_	[20]
GA	Ep	54.0	_	150.0	_	_	_	50.0	_	140.0	[27]
AF	PE	200.0	_	350.0	_	_	_	_	_	_	[28]
SS	Cu	_	_	_	_	_	_	1,700	200	1,000	[48]

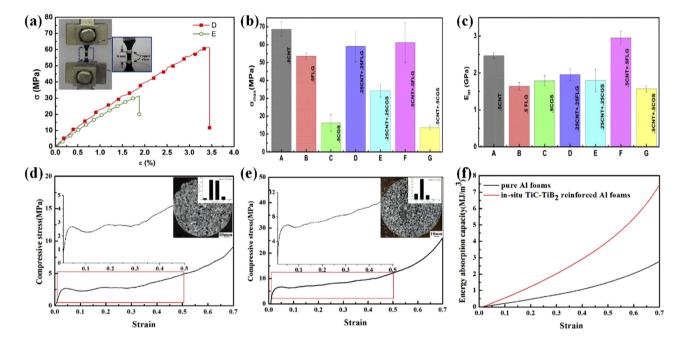


Figure 5: Mechanical properties of the nanocomposites. (a) Stress-strain curves for two selected materials of D (0.25 CNT + 0.25 FLG) and E (0.25 CNT + 0.25 CGS), (b) elastic modulus, and (c) tensile strength [54] (Copyright 2018, Elsevier). Compressive stress-strain curves and energy absorption capacity curves of pure Al foam and *in situ* TiC-TiB₂-reinforced Al foam: (d) and (e) compressive stress-strain curves and pore structures (inset of d and e), and (f) energy absorption capacity curves [57] (Copyright 2020, Elsevier).

off or cracking appears between the composite layer and core. Tilbrook studied the mechanical properties and fatigue characteristics of AF/EP composites and found that such interpenetrating phase composites (IPCs) are more excellent [51]. Similarly, Dukhan *et al.* studied AF/PP-IPCs and found that the smaller the AF pore size, the greater the bending strength of the composite material, and the mathematical relationship between structural parameters and bending properties was established [30]. Yin from the same group used a similar method to prepare the composite materials. The filling phases he

used were silicone rubber and room temperature-vulcanized rubber to study the damping properties of the composite materials [52]. Liu and Gong prepared AF/PE-IPCs and AF/EP-IPCs, and compared their static compression and energy absorption properties with pure aluminum and AF, and found that AL/EP-IPCs are materials with better absorption properties [28]. Liu studied the cushioning performance of AF/polyurethane-IPCs through finite element simulation and found that it has good cushioning performance [53].

In addition, Avilés *et al.* studied the mechanical properties of thermosetting polymer nanocomposites comprising

carbon nanotubes (CNTs, one-dimensional topology, 1D), few-layer thermally reduced graphene oxide (FLG, twodimensional topology, 2D), cubic-shaped few-layer graphene shells (CGSs, three-dimensional topology, 3D), and hybrid combinations of them (1D-2D and 1D-3D). The stress-strain curve of all nanocomposite materials exhibits brittle behavior of thermoset material and the mechanical properties of the D is the highest (Figure 5a) [54]. By comparing the mechanical properties of composites fabricated with only one filler (Figure 5b and c), it is seen that those with CNTs have a better mechanical performance, followed by those made from FLGs, which indicate that this topological combination and the high density of functional groups in the sheet are beneficial to the stress transfer inside the composite. Hou studied the reliability of auxetic (re-entrant honeycomb) and nonauxetic (diamond lattice and conventional honeycomb) lattice composites and the findings pave the way for developing new class of auxetic lattice composites, especially under cyclic loading conditions, through a combination of rational design and 3D printing [55]. In addition, polymers can be used to strengthen the aluminum foams to make composites. Recently, the Al composite foams reinforced by ceramic particles have been developed due to the great potential of ceramic particles in improving the mechanical properties of Al foam [56]. An et al. successfully fabricated in situ TiC-TiB-reinforced AF with excellent mechanical properties via a novel method by the combination of Ti-BC self-propagating reaction and melt processing technique [57]. The results showed that the interfacial bonding of TiB/Al and TiC/Al was excellent and the foams exhibited a superior mechanical property and energy absorption capacity as well as uniform deformation simultaneously (inset of Figure 5d and e). Compared with pure Al foams, the compressive strength (Figure 5d and e) and energy absorption capacity (Figure 5f) of the in situ TiC-TiB-reinforced Al foams improved by about 149 and 168%, respectively. Glass fibers are usually used as enhanced materials, electrically insulating materials, thermally insulating materials, circuit boards, and in other fields. Tian's group studied the effect of adding hollow glass beads on the compression and bending properties of AF/EP composites, and found that as the volume content of hollow glass beads increase, the mechanical properties of the composites gradually decrease [18]. Jhaver and Tippur prepared two kinds of hollow glass beads with different volume fractions and IPCs formed by the composite foam formed by EP and AF composited, which reached their effective elastic modulus and yield stress, and the results show that

the performance of composite materials is better than that of syntactic foam [58].

While conducting experimental research on the mechanical properties of the composite materials, it is also necessary to explain them theoretically. Regarding the enhancement mechanism of multiphase materials, various scholars have proposed a variety of models since the 1870s. The most classic model is the "Hashin-Shtrikman" model established by Hashin and Shtrikman [60]. The establishment of this model fills the gaps in the research on the enhancement mechanism of network interweaving. explains the enhancement mechanism of the network structure to the composite material in a more scientific way, and describes more accurately the changes in the modulus and strength of the material after enhancement, and can give the theoretical modulus and theoretical strength range of the composite materials [61]. The more classic calculation process is as follows (twophase material):

The two phases in the composite material penetrate each other and each are continuous. The mass of the material can be calculated by formula (1):

$$\rho V = \sum \rho_i V_i, \qquad (1)$$

where ρ and V are the density and volume of the material, respectively, and i is the number of phases.

Defining the bulk elastic modulus of the low modulus phase material as K_1 , the shear modulus as G_1 , and the volume fraction as V_1 and the bulk elastic modulus of the high modulus phase material as K_2 , the shear modulus as G_2 , and the volume fraction as V_2 , the effective volume modulus range $K_L - K_H$ can be obtained by the following formula:

$$K_{\rm L} = K_1 + \frac{V_2}{\frac{1}{K_2 - K_1} + \frac{3V_1}{3K_1 + 4G_1}},$$
 (2)

$$K_{\rm H} = K_2 + \frac{V_1}{\frac{1}{K_1 - K_2} + \frac{3V_2}{3K_2 + 4G_2}}$$
 (3)

Similarly, the prediction formulas of the shear modulus range are:

$$G_{\rm L} = G_1 + \frac{V_2}{\frac{1}{G_2 - G_1} + \frac{6V_1(K_1 + 2G_1)}{5G_1(3K_1 + 4G_1)}},\tag{4}$$

$$G_{\rm H} = G_2 + \frac{V_1}{\frac{1}{G_1 - G_2} + \frac{6V_2(K_2 + 2G_2)}{5G_2(3K_2 + 4G_2)}}.$$
 (5)

From this, the predicted range of the Young's modulus of the material can be calculated as:

$$(E_{\rm L} - E_{\rm H}) : E_{\rm L} = \frac{9K_{\rm L}G_{\rm L}}{3K_{\rm L} + G_{\rm L}},$$
 (6)

$$E_{\rm H} = \frac{9K_{\rm H}G_{\rm H}}{3K_{\rm H} + G_{\rm H}}. (7)$$

However, with the further development of research, people have found that the Hashin–Shtrikman model has many shortcomings and its scope of application is also limited. Later Tuchinskii model [62], the Gurson–Tvergaard–Needleman model [63] and others are more focused on the interpretation of single problems such as unified network structure and flexible failure of composite materials, which can more accurately reveal the mechanical behavior of the composite materials [64]. For example, Basirat *et al.* used a continuous failure model to simulate and calculate the creep behavior of manganese-chromium 3D-NTSC [65].

On the basis of numerically predicting the strength and modulus of the composite materials with mathematical models, the mechanical behavior of composite materials can be simulated by computer, which can observe and record the behavior changes of composite materials in real time. Swanson developed a finite element simulation program, which opened the history of simulation and with the help of simulation software, loading the force field, you can get the behavior change of the material in the force process [66,67]. While studying the mechanical properties of AF-based 3D-NTSC, Hong et al. used finite element analysis methods to scientifically and accurately predict the failure mode of the composite materials during compression [23]. A mathematical model of the compressive plateau stress of the AF/EP composite material was established by using the "mixing ratio" and "synergistic effect" of the composite material. Compared with the experimental results and finite element simulation results [68], the deformation and failure processes of the material during compression and the variation of stress-strain relationship with the structural parameters and strain rates are in good agreement [69]. Jhaver and Tippur used finite element analysis to obtain the force mode and deformation of the cell network of the composite material during compression [58]. Dukhan used finite element analysis to analyze the deformation mode of the composite materials under tensile stress and so on [70].

5 Summary and outlook

Throughout the related research of 3D-NTSC, we can see that the performance and theoretical research of 3D-NTSC

have been developed quite maturely, and people have been able to successfully predict the modulus and strength of composite materials theoretically. The increase in the strength of composite materials is conducive to expanding the application range of the composite materials and demonstrating the functionality of the composite materials. Based on the advantages of single-phase materials, composite materials have excellent properties in electrical conductivity [71], thermal conductivity [72,73], wave absorption [74,75], and damping [76,77].

However, in fact, the application of 3D-NTSC in practice is not very broad, the main reason is the lack of preparation and performance. In preparation, if the materials are to be well infiltrated, either to improve the interface properties or to remove the interface air or apply external force, this will increase the actual production cost and the process will be more complicated. The mechanical properties of the reinforced materials cannot be significantly improved under the condition of high-performance retention. To a certain extent, it can be considered that the filling reinforcement is the damage to the original structure. Therefore, the following prospects are put forward regarding the development of 3D-NTSC:

- (1) Try simpler processing techniques to prepare 3D-NTSC, such as hot pressing.
- (2) Establish the matching relationship between the mechanical properties of the matrix phase and the filling phase, and optimize the materials of each phase through theoretical calculations in the early stage, and obtain composite materials with excellent comprehensive properties as much as possible without damaging the characteristics of single-phase materials.
- (3) The optimization of the properties of composite materials is achieved by optimizing the properties and structural parameters of the network structure. The main influencing factor in this respect is that the quality of raw materials cannot be guaranteed in our country at present.

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References

- [1] Al-Ketan O, Abu Al-Rub RK, Rowshan R. Mechanical properties of a new type of architected interpenetrating phase composite materials. Adv Mater Technol. 2017;2:1600235.
- Fan ZQ, Zhang BB, Liu YB, Suo T, Xu P, Zhang JJ. Interpenetrating phase composite foam based on porous aluminum skeleton for high energy absorption. Polym Test. 2021;93:106917.
- [3] Li TT, Chen YY, Wang LF. Enhanced fracture toughness-in architected-interpenetrating phase composites by 3D printing. Compos Sci Technol. 2018;167:251-9.
- Lin YZ, Zhong LB, Dou S, Shao ZD, Liu Q, Zheng YM. Facile synthesis of electrospun carbon nanofiber/graphene oxide composite aerogels for high efficiency oils absorption. Environ Int. 2019;128:37-45.
- Zhang Z, Luo SB, Yu SH, Guan ZS, Sun R, Wong CP. Significantly enhanced dielectric and energy storage performance of blend polymer-based composites containing inorganic 3D-network. Mater Des. 2018;142:106-13.
- Aldrich DE, Fan Z. Microstructural characterization of interpenetrating nickel/alumina composites. Mater Charact. 2001;47:167-73.
- Goller G, Koty DP, Tewari SN, Singh M, Tekin A. Wear and friction behavior of metal impregnated microporous carbon composites. Metall Mater Trans A Phys Metall Mater Sci. 1996;27:3727-38.
- [8] Al-Ketan O, Assad MA, Abu Al-Ru RK. Mechanical properties of periodic interpenetrating phase composites with novel architected microstructures. Compos Struct. 2017;176:9-19.
- Jurewicz I, Worajittiphon P, King AAK, Sellin PJ, Keddie JL, Dalton AB. Locking carbon nanotubes in confined lattice geometries-a route to low percolation in conducting composites. J Phys Chem B. 2011;115:6395-400.
- [10] Li JQ, Wang YY, Ling H, Qiu Y, Lou J, Hou X, et al. Significant enhancement of the visible light photocatalytic properties in 3D BiFeO₃/graphene composites. Nanomaterials. 2019;9:1.
- [11] Biggemann J, Diepold B, Pezoldt M, Stumpf M, Greil P, Fey T. Automated 3D assembly of periodic alumina-epoxy composite structures. J Am Ceram Soc. 2018;101:3864-73.
- [12] Guarino V, Ambrosio L. The synergic effect of polylactide fiber and calcium phosphate particle reinforcement in poly epsiloncaprolactone-based composite scaffolds. Acta Biomater. 2008;4:1778-7.
- [13] Tong L, Mouritz AP, Bannister M. 3D Fibre reinforced polymer composites. Oxford: Elsevier Science; 2002. p. 219-36.
- [14] Challagulla KS, Georgiades A, Kalamkarov AL. Asymptotic homogenization model for three-dimensional network reinforced composite structures. J Mech Mater Struct. 2007;2:613-32.
- [15] Tan Y, Qian WP, Ding SH, Wang Y. Gold-nanoparticle-infiltrated polystyrene inverse opals: a three-dimensional platform for generating combined optical properties. Chem Mater. 2006;18:3385-9.

- [16] Kang YL, Yu FR, Chen C, Zhang Q, Sun H, Zhang L, et al. High pressure synthesis and thermoelectric properties of Ba-filled CoSb₃ skutterudites. J Mater Sci Mater Electron. 2017;28:8771-6.
- [17] Liu WW, Lu CX, Wang XL, Liang K, Tay BK. In situ fabrication of three-dimensional, ultrathin graphite/carbon nanotube/NO composite as binder-free electrode for high-performance energy storage. J Mater Chem A. 2015;3:624-33.
- [18] Tian A, Yu W, Li D. Compressive and flexural mechanical properties of foam Al-Hgm/epoxy foam composites. Acta Mater Compos Sin. 2013;30:74-81.
- [19] Liu JY, Ping XU, Ying-Hua YU. Manufacturing and damping properties of the composite material of foam aluminum. Coal Mine Mach. 2004:5:33-5.
- [20] Yu W, Li H, Zhao Z, Liang X. Compressive mechanical properties of foam aluminum-epoxy interpenetrating phase composites. Acta Mater Compos Sin. 2012;29:224-30.
- [21] Breslin MC. Transformation Kinetics of Al₂O₃/Al Co-continuous ceramic/metal composite materials (C4) produced by a displacement reaction between liquid Al and fused SiO2. Ohio: The Ohio State University; 1994.
- [22] Aghajanian MK, Burke JT, White DR, Nagelberg AS. A new infiltration process for the fabrication of metal-matrix. Composites. 1989;20:43-6.
- [23] Hong CQ, Han HC, Zhang XH, Meng SH, Li JP. Ablation-resistance behavior of sweat cooling Tib2-Cu matrix interpenetrating phase composites. Rare Met Mater Eng. 2007;36:546-9.
- Chen XW, Dong SM, Kan YM, Jin XH, Zhou HJ, Ni DW, et al. Microstructure and mechanical properties of three dimensional C_f/SiC-ZrC-ZrB₂ composites prepared by reactive melt infiltration method. J Eur Ceram Soc. 2016;36:3969-76.
- [25] Liang LY, Li QM, Yan X, Feng YZ, Wang YM, Zhang HB, et al. Multifunctional magnetic Ti₃C₂Tx MXene/graphene aerogel with superior electromagnetic wave absorption performance. ACS Nano. 2021;15:6622-32.
- [26] Li CW, Jiang DG, Liang H, Huo BB, Liu CY, Yang WR, et al. Superelastic and arbitrary-shaped graphene aerogels with sacrificial skeleton of melamine foam for varied applications. Adv Funct Mater. 2018;28(8):1704674.1-9.
- [27] Ni Y, Chen L, Teng KY, Shi J, Qian XM, Xu ZW, et al. Superior mechanical properties of epoxy composites reinforced by 3D interconnected graphene skeleton. ACS Appl Mater Interfaces. 2015;7:11583-91.
- [28] Liu Y, Gong XL. Compressive behavior and energy absorption of metal porous polymer composite with interpenetrating network structure. Trans Nonferrous Met Soc China. 2006;16:439-43.
- [29] Fang XD, Bi C, Hong YF, Cho KH, Park MS, Wang YJ, et al. Rapid vacuum infusion and curing of epoxy composites with a rubber-cushioned mold design. Polym Technol Eng. 2016;55:1030-8.
- [30] Dukhan N, Rayess N, Hadley J. Characterization of aluminum foam-polypropylene interpenetrating phase composites: flexural test results. Mech Mater. 2010;42:134-41.
- Long CG, Zhang Z, Guo FY, Huang JL, Cao TS. Effect of different porosity on aluminum alloy foam/polyformaldehyde interpenetrating phase composites. J Changsha Univ Sci Technol (Nat Sci). 2014;3:88-94.
- [32] Long CG, Su Y, Shen C. Mechanical and tribological properties of polytetrafluoroethylene filled polyoxymethylene/aluminum

- foam interpenetrating phase composites. Adv Mater Res. 2014;835-836:285-9.
- [33] Su Y, Long CG, Zhou Y, Cao TS, Hua MY. New aluminum foam/ polyformaldehyde interpenetrating phase composites prepared by injection molding. Adv Mater Res. 2013;631-632:13-7.
- [34] Kocaba M, Rnek C, Curioni M, Cansever N. Nickel fluoride as a surface activation agent for electroless nickel coating of anodized AA1050 aluminum alloy. Surf Coat Technol. 2019;364:231-8.
- [35] Zhang XH, Zhang Y, Tian BH, Jia YL, Liu Y, Song KX, et al. Cr effects on the electrical contact properties of the Al₂O₃-Cu/ 15W composites. Nanotechnol Rev. 2019;8:128-35.
- [36] Vinogradov NA, Harlow GS, Carla F, Evertsson J, Rullik L, Linpe W, et al. Observation of pore growth and self-organization in anodic alumina by time-resolved X-ray scattering. ACS Appl Nano Mater. 2018;1:1265-71.
- [37] Larsson A, Abbondanza G, Linpe W, Carla F, Mousley P, Hetherington C, et al. Electrochemical fabrication and characterization of palladium nanowires in nanoporous alumina templates. J Electrochem Soc. 2020;167(12):122514 (10pp).
- [38] Liu YS, Meng FB, Huang JC, Ni LJ, Shen YD, Zhang LY. Polyphenylene sulfide-coated wrench composites by nanopinning effect. Nanotechnol Rev. 2021;10:166-77.
- [39] Feng HJ, Moore JJ. In situ combustion synthesis of dense ceramic and ceramic-metal interpenetrating phase composites. Metall Mater Trans B Process Metall Mater Process Sci. 1995;26:265-73.
- [40] Fan Z, Gong F, Nguyen ST, Duong HM. Advanced multifunctional graphene aerogel-poly(methyl methacrylate) composites: experiments and modeling. Carbon. 2015;81:396-404.
- [41] Aldosari MA, Othman AA, Alsharaeh EH. Synthesis and characterization of the in situ bulk polymerization of PMMA containing graphene sheets using microwave irradiation. Molecules. 2013;18:3152-67.
- [42] Peroni L, Avalle M, Peroni M. The mechanical behaviour of aluminium foam structures in different loading conditions. Int J Impact Eng. 2008;35:644-58.
- [43] Ma JX, Li WH, Fan YC, Yang JY, Yang QK, Wang JC, et al. Ultrathin and light-weight graphene aerogel with precisely tunable density for highly efficient microwave absorbing. ACS Appl Mater Interfaces. 2019;11:46386-96.
- [44] Cao XY, Zhang J, Chen SW, Varley RJ, Pan K. 1D/2D Nanomaterials synergistic, compressible, and response rapidly 3D graphene aerogel for piezoresistive sensor. Adv Funct Mater. 2020;30:2003618.
- [45] Wang X, Yu ZH, Bian HY, Wu WB, Xiao HN, Dai HQ. Thermally conductive and electrical insulation BNNS/CNF aerogel nanopaper. Polymers. 2019;11:660.
- [46] Si Y, Wang LH, Wang XQ, Tang N, Yu JY, Ding B. Ultrahighwater-content, superelastic, and shape-memory nanofiberassembled hydrogels exhibiting pressure-responsive conductivity. Adv Mater. 2017;29:1700339.
- [47] Zhukhovitskiy AV, Zhong MZ, Keeler EG, Michaelis VK, Sun JEP, Hore MJA, et al. Highly branched and loop-rich gels via formation of metal-organic cages linked by polymers. Nat Chem.
- [48] Yuan ZY, Rayess N, Dukhan N. Modeling of the mechanical properties of a polymer-metal foam hybrid. In: Rabiei A, editor.

- 8th International Conference on Porous Metals and Metallic Foams; 2014. p. 215-9
- [49] Chen SJ, Ren DX, Li B, Xu MZ, Liu XB. Investigation on phenolphthalein and bisphenol AF based poly(arylene ether nitrile) copolymers: preparation, thermal, mechanical and dielectric properties. Polym Test. 2021;96:107091.
- [50] Cheng SL, Zhao XY, Liu XM, Yu W, Xin YJ. Flexural behavior of composite sandwich panel of aluminum foam and epoxy resin. Emerg Mater Res. 2017;6:276-84.
- [51] Tilbrook MT, Moon R, Hoffman M. On the mechanical properties of alumina-epoxy composites with an interpenetrating network structure. Mater Sci Eng A Struct Mater Prop Microstruct Process. 2005;393:170-8.
- [52] Yin SJ, Rayess N. Characterization of polymer-metal foam hybrids for use in vibration dampening and isolation. Proc Mater Sci. 2014;4:311-6.
- [53] Liu SB, Li AQ, Xuan P. Mechanical behavior of aluminum foam/ polyurethane interpenetrating phase composites under monotonic and cyclic compression. Compos Part A Appl Sci Manuf. 2019;116:87-97.
- [54] Aviles F, May-Pat A, Lopez-Manchado MA, Verdejo R, Bachmatiuk A, Rummeli MH. A Comparative study on the mechanical, electrical and piezoresistive properties of polymer composites using carbon nanostructures of different topology. Eur Polym J. 2018;99:394-402.
- [55] Hou SY, Li TT, Jia Z, Wang LF. Mechanical properties of sandwich composites with 3D-printed auxetic and non-auxetic lattice cores under low velocity impact. Mater Des. 2018;160:1305-21.
- [56] Duarte I, Ferreira JMF. Composite and nanocomposite metal foams. Materials. 2016;9:79.
- [57] An XG, Liu Y, Ye JW, Li X. Fabrication of in-situ TiC-Tib2 reinforced al foam with enhanced properties. J Alloy Compd. 2020;825:153969.
- [58] Jhaver R, Tippur HV. Processing, compression response and finite element modeling of syntactic foam based interpenetrating phase composite (IPC). Mater Sci Eng A Struct Mater Prop Microstruct Process. 2009;499:507-17.
- [59] Okulov IV, Weissmuller J, Markmann J. Dealloying-based interpenetrating-phase nanocomposites matching the elastic behavior of human bone. Sci Rep. 2017;7:20.
- [60] Hashin Z, Shtrikman S, Solids P. A variational approach to the theory of the elastic behaviour of multiphase material. J Mech Phys Solids. 1963;11:127-40.
- [61] Watt JP. Hashin-Shtrikman bounds on the effective elastic moduli of polycrystals with irigonal $(3, \bar{3})$ and tetragonal $(4, \bar{4})$ 4 m) symmetry. J Appl Phys. 1986;60:3120-4.
- [62] Tuchinskii L. Elastic constants of pseudoalloys with a skeletal structure. Sov Powder Metall Met Ceram. 1983;22:588-95.
- [63] Gurson AL. Continuum theory of ductile rupture by void nucleation and growth: part i-yield criteria and flow rules for porous ductile media. J Eng Mater Technol. 1977;99:297-300.
- [64] Okulov I, Weissmüller J, Markmann J. Dealloying-based interpenetrating-phase nano composites matching the elastic behavior of human bone. Sci Rep. 2017;7:1-7.
- [65] Basirat M, Shrestha T, Potirniche GP, Charit I, Rink K. A study of the creep behavior of modified 9Cr-1MO steel using continuum-damage modeling. Int J Plasticity. 2012;37:95-107.

- [66] Swanson JA, Cameron GR, Haberland JC. Adapting the ansys finite-element analysis program to an attached processor. Computer. 1983;16:85-91.
- [67] Swanson JA. Keynote paper: current and future trends in finite element analysis. Int J Comput Appl Technol. 1994;7:108-17.
- [68] Dalaq AS, Abueidda DW, Abu Al-Rub RK, Jasiuk IM. Finite element prediction of effective elastic properties of interpenetrating phase composites with architectured 3D sheet reinforcements. Int J Solids Struct. 2016;83:169-82.
- [69] Yu YH, Bing L. Calculation of effective elastic modulus of foamed aluminum/epoxy resin composite based on IPC material. Mater Mech Eng. 2008;32:90-2.
- [70] Dukhan N. Metal foams: fundamentals and applications. Appl Therm Eng. 2013:61:1.
- [71] Bi H, Lin TQ, Xu F, Tang YF, Liu ZQ, Huang FQ. New graphene form of nanoporous monolith for excellent energy storage. Nano Lett. 2016;16:349-54.
- [72] Miltz J, Gruenbaum G. Evaluation of cushioning properties of plastic foams from compressive measurements. Polym Eng Sci. 1981;21:1010-4.

- [73] Evans AG, Hutchinson J, Ashby MF. Multifunctionality of cellular metal systems. Prog Mater Sci. 1998;43:171-221.
- Farhan S, Wang RM, Li KZ. Carbon foam decorated with silver particles and in situ grown nanowires for effective electromagnetic interference shielding. J Mater Sci. 2016;51:7991-8004.
- [75] Al-Ghamdi AA, Al-Ghamdi AA, Al-Turki Y, Yakuphanoglu F, El-Tantawy F. Electromagnetic shielding properties of graphene/acrylonitrile butadiene rubber nanocomposites for portable and flexible electronic devices. Compos Part B Eng. 2016;88:212-9.
- [76] Chen SB, Wang TM, Wang QH. Damping properties of polyurethane/epoxy graft interpenetrating polymer network composites filled with short carbon fiber and nano-SiO2. J Macromol Sci Part B Phys. 2011;50:931-41.
- [77] Wang TM, Chen SB, Wang QH, Pei XQ. Damping analysis of polyurethane/epoxy graft interpenetrating polymer network composites filled with short carbon fiber and micro hollow glass bead. Mater Des. 2010;31:3810-5.