



## Editorial/preface

## Conductive polymer composites with segregated structures



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## ABSTRACT

Conductive polymer composites (CPCs) have generated significant academic and industrial interest for several decades. Unfortunately, ordinary CPCs with random conductive networks generally require high conductive filler loadings at the insulator/conductor transition, requiring complex processing and exhibiting inferior mechanical properties and low economic affordability. Segregated CPC (s-CPC) contains conductive fillers that are segregated in the perimeters of the polymeric granules instead of being randomly distributed throughout the bulk CPC material; these materials are overwhelmingly superior compared to normal CPCs. For example, the s-CPC materials have an ultralow percolation concentration (0.005–0.1 vol%), superior electrical conductivity (up to  $10^6$  S/m), and reasonable electromagnetic interference (EMI) shielding effectiveness (above 20 dB) at low filler loadings. Therefore, considerable progress has been achieved with s-CPCs, including high-performance anti-static, EMI shielding and sensing materials. Currently, however, few systematic reviews summarizing these advances with s-CPCs are available. To understand and efficiently harness the abilities of s-CPCs, we attempted to review the major advances available in the literature. This review begins with a concise and general background on the morphology and fabrication methods of s-CPCs. Next, we investigate the ultralow percolation behaviors of and the elements exerting a relevant influence (e.g., conductive filler type, host polymers, dispersion methods, etc.) on s-CPCs. Moreover, we also briefly discussed the latest advances in the mechanical, sensing, thermoelectric and EMI shielding properties of the s-CPCs. Finally, an overview of the current challenges and tasks of s-CPC materials is provided to guide the future development of these promising materials.

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**Abbreviations:** 1D, one-dimensional; 2D, two-dimensional; 3D, three-dimensional; AAEM, acetoacetoxyethyl methacrylate; ABS, acrylonitrile–butadiene–styrene; AFM, atomic force microscopy; BA, butyl acrylate; CB, carbon black; CF, carbon fiber; CNT, carbon nanotube; CPC, conductive polymer composite; CuNW, copper nanowire; EG, expanded graphite; EMI, electromagnetic interference; EMI SE, electromagnetic interference shielding effectiveness; GA, Gum Arabic; GNS, graphene nanosheet; HDPE, high-density polyethylene; HIPS, high-impact polystyrene; IT, isothermal treatment; ITO, indium tin oxide; ISI, Institute for Scientific Information; LDPE, low-density polyethylene; MA, maleic anhydride; MAA, methacrylic acid; MMA, methyl methacrylate; MWNT, multi-wall carbon nanotube; MSMP, miscible solvent mixing and precipitation; MWD, molecular weight distribution; NR, natural rubber; NTC, negative temperature coefficient; PA, polyamide; PANI, polyaniline; PC, polycarbonate; PE, polyethylene; PEDOT:PSS, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate); PET, poly(ethylene terephthalate); PMMA, poly(methyl methacrylate); PP, polypropylene; PPS, poly(phenylene sulfide); PS, polystyrene; PTC, positive temperature coefficient; PVAc, poly(vinyl acetate); PVC, poly(vinyl chloride); PVDF, poly(vinylidene fluoride); SAN, poly(styrene-co-acrylonitrile); s-CPC, segregated conductive polymer composite; SDS, sodium dodecyl sulfate; SEM, scanning electron microscopy; SWNT, single-wall carbon nanotube;  $T_g$ , glass transition temperature; TCPP, *meso*-tetra(4-carboxyphenyl) porphine; UHMWPE, ultrahigh molecular weight polyethylene; WPU, waterborne polyurethane; ZT, thermoelectric figure of merit.

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## 1. Introduction

Electrically conductive polymer composites (CPCs), which consist of single or hybrid conductive fillers (e.g., carbonaceous, metallic, and conducting polymeric particles) dispersed in a polymer matrix based on a single polymer or a multi-phase blend, have attracted considerable academic and industrial attention for several decades [1–5]. Their popularity can be demonstrated by the amount of research articles relevant to CPCs: over 12,000 publications was retrieved on 20 May, 2014 on the Institute for Scientific Information (ISI)—Web of Science database, searching the subject “conductive polymer composite”. Owing to their ease of processing, low-cost, and tunable electrical properties in comparison to intrinsic conducting polymers, CPCs have served applications as anti-static materials, electromagnetic interference (EMI) shielding, sensors and conductors [6–9]. The electrical resistivity of CPCs determines their specific applications, e.g., see Fig. 1. For example, the CPC materials for electrostatic dissipation in plastic fuel tanks typically require an electrical resistivity of  $\sim 10^6 \Omega \text{ cm}$ , while EMI shielding requires electrical resistivity values below  $10^{-2} \Omega \text{ cm}$ .

Because most of the ordinary host polymers are essentially insulating, the electrical performance of CPCs relies solely on continuous conductive networks constructed after incorporating the conductive fillers [10–12]. When the conductive filler content reaches a critical value, the CPC material will exhibit an insulator/conductor transition; specifically, the electrical conductivity dramatically increases by several orders of magnitude when the initial conducting channels are formed. This critical volume fraction  $\phi$  is defined as the percolation threshold  $\phi_c$ . As the conductive filler content increases, additional conductive pathways may be established in the polymer matrix, allowing the electrical conductivity to increase gradually until saturation plateau is reached.

The electrically conductive behavior of a CPC material may usually be empirically described the power law [11,12]:

$$\sigma = \sigma_0(\phi - \phi_c)^t \quad (1)$$

where  $\sigma$  represents the CPC electrical conductivity and  $t$  is the critical exponent related to the dimensionality of the conductive networks in CPC. In this model,  $t \approx 2$  and  $t \approx 1.3$  are for three-dimensional (3D) and two-dimensional (2D) conductive networks, respectively. However, the experimental values of usually deviate from these predicted values [13,14].

Among the conventional CPC fabrication methods (i.e., solution processing, melt mixing, and in situ polymerization) [1,2,4], the melt mixing technologies, such as twin-screw extrusion, internal mixing and injection molding, are the most common approaches used to fabricate commercial CPC materials because these techniques are compatible with current industrial practices. Nonetheless, CPCs fabricated via conventional melt-mixing approaches generally have a relatively high  $\phi_c$ . Theoretically, the  $\phi_c$  for spherical, randomly dispersed fillers (e.g., carbon black (CB), metallic particles, and conducting polymer particles) is approximately 10–20 vol%, which is close to the percolation value of  $\sim 16 \text{ vol\%}$  predicted by the classical percolation theory [15,16]. Although high-aspect-ratio conductive nano-particles (e.g., carbon nanotube (CNT) or graphene nanosheet (GNS)) have large surface areas able to support well-developed transport networks, their extreme agglomeration behavior during processing in host polymers generates the relatively high  $\phi_c$ . Unfortunately, the CPCs with high  $\phi_c$  always suffer from various drawbacks: (i) high melt viscosities, (ii) low economic affordability, and (iii) inferior mechanical properties (esp. for ductility and toughness) [17,18]. Therefore, decreasing  $\phi_c$  efficiently has become a long-standing, major topic during the fabrication of high-performance CPC materials.

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