Accepted Manuscript


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PII: S1001-8417(17)30304-2
DOI: http://dx.doi.org/10.1016/j.cclet.2017.08.020
Reference: CCLET 4169

To appear in: *Chinese Chemical Letters*

Please cite this article as: Jinkai Yuan, Percolation of carbon nanomaterials for high-k polymer nanocomposites, Chinese Chemical Lettershttp://dx.doi.org/10.1016/j.cclet.2017.08.020

Review

# Percolation of carbon nanomaterials for high-*k* polymer nanocomposites

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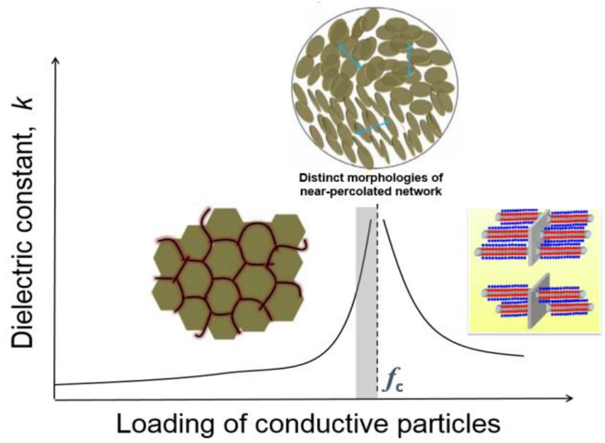
Graphical abstract




This review summarized the recent progress towards high-*k* polymer composites bases on the near-percolated networks of carbon nanomaterials by focusing on the effects of distinct network morphologies on the dielectric properties. It is expected to give guidance on designing new near-percolated networks in polymer matrices towards next-generation polymer dielectrics.

*ARTICLE INFO: Article history*, Received 27 June 2017, Received in revised form 1 August 2017, Accepted 3 August 2017, Available online

*ABSTRACT*
*High-*k *polymer composite materials are next-generation dielectrics that show amazing applications in diverse electrical and electronic devices. Establishing near-percolated network of conducting filler in an insulating polymer matrix is a promising approach to develop flexible high-*k *dielectrics. However, challenges still exist today on fine controlling the network morphology to achieve extremely high* k *values and low losses simultaneously. The relationship between the network morphology and the dielectric properties of polymer composites is raising a number of fundamental questions. Herein, recent progress towards high-*k *polymer composites based on carbon nanomaterials is reviewed. Particular attention is paid on the influence of the network morphology on the dielectric properties. Some perspectives that warrant further investigation in the future are also addressed*

Keywords:Percolation Carbon nanomaterials Polymer composites Dielectric constant Permittivity

## 1. Introduction

Over the past decades, dielectric polymers are rapidly emerging as novel materials for diverse engineering applications, such as energy storage and conversion [1-6], electrocaloric cooling [7, 8], actuators [9-11], and sensors [12-14], since they have advantages in high voltage rating and processing ease over conventional ceramic dielectrics. Unfortunately, most of the commercial polymers suffer from limited dielectric constant ($2<k<5$) due to the electronic and atomic polarization of hydrocarbon covalent bonds [5], and thereby fall short of the rising demands for the higher functionality and further miniaturization of next-generation electronic and electrical devices. The key to address this issue is to substantially raise the dielectric constant of polymers, meanwhile retaining their other inherent properties, such as high breakdown strength and low dielectric losses.

The introduction of high-*k* nanofillers into polymer matrices to form nanocomposite dielectrics represents a straightforward avenue to this end [15-17]. The rationale behind this approach lies in the combination of the impressive dielectric constant of filler particles and the high dielectric strength and low dielectric losses of polymer matrices. In such way, the high-*k* nanoparticles contribute to the increment of the effective dielectric constant of nanocomposites in terms of the effective medium theory. Previous studies validate that the dielectric properties can be fine tailored by rationally selecting the shape and size of high-*k* fillers as well as modulating the anisotropy or topological structure of the nanocomposites [18-20]. Indeed, numerous research has succeeded in improving the overall dielectric performances using 0-D metal oxide nanoparticles such as copper calcium titanate (CCTO) [21], 1-D perovskite fibers such