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Review

Polymer composite hydrogels containing carbon nanomaterials—Morphology and mechanical and functional performance

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ABSTRACT

Carbon nanomaterials have been the centre of intensive research because of their unique structure and technologically useful properties. We herein review the effect of the nanomaterials on the morphology and mechanical and functional properties of composite hydrogels, by critically examining the nanomaterial dispersion in aqueous media and their interaction with polymer matrices. We focus on synthesis methods and the mechanical, electrical and swelling properties of the hydrogels with emphasis on cross-linking techniques and structure-property relations. The nanomaterial properties and fabrication techniques are briefly discussed highlighting advantages and limitations. This review outlines the current challenges and opportunities to effectively utilizing carbon nanomaterials for polymer hydrogels. The output of such materials would be valuable to soft materials scientists and composite engineers, as well as those working on water-soluble, synthetic polymers.

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Nomenclature

Acronyms

ATRP	Atom transfer radical polymerization
BIS	*N*,*N*-methylenebisacrylamide
CNTs	Carbon nanotubes
FRP	Free-radical polymerization
GnPs	Graphene platelets
GO	Graphene oxide
MWCNT	Multi walled carbon nanotubes
NC	Polymer nanocomposite
OR	Organic crosslinker
PAA	Poly(acrylic acid)
PAAG	Poly(acrylic acid)-*g*-gelatin
PAM	Polyacrylamide
PNIPAM	Poly(*N*-isopropyl acrylamide)
PPy	Polypyrrole
PVA	Poly(vinyl alcohol)
PVP	Poly(vinyl pyrrolidone)
RGO	Reduced graphene oxide
SEM	Scanning electron microscopy
SWCNTs	Single walled carbon nanotubes
RAFT	Reversible addition-fragmentation chain transfer polymerization
TMEDA	Tetramethylethylenediamine

1. Introduction

Of all engineering materials, polymers have seen the greatest increase in applications over the past decades, owing to their high ratio of strength to density, ease of processing and relatively low energy consumption required in manufacturing [1]. However, most polymers lack sufficient mechanical strength and functionality, in specific electrical and thermal conductivity [2]. To overcome these limitations, more than 95% of polymers are compounded with organic/inorganic additives in manufacturing to create composites [3]. However, these attempts are only partially successful, because the composites have *micron*-sized dispersion phases; their mechanical properties sometimes fall significantly short of the needs of many applications that require a nano-sized dispersion phase [4].

Polymer composites are produced by compounding fillers with polymers to obtain markedly improved or new properties beyond their individual components. Soft and wet materials, such as hydrogels, are liquid-swollen polymeric materials with distinct 2D or 3D structures formed by chemical or physical cross-linking [5]. Of various gels, chemically cross-linked polymer hydrogels are the most widely used in applications. In principle composite hydrogels based on carbon nanomaterials should combine the merits of the nanomaterials with those of the hydrogels. An emerging research area is the development of new electrically conductive, mechanically robust composite hydrogels for energy storage and bioengineering applications [6–9].

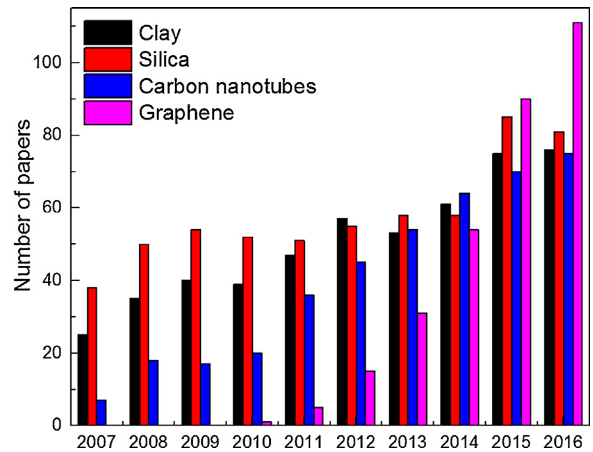

Fig. 1. Publications for composite hydrogels respectively containing clay, silica, carbon nanotubes and graphene. (Web of Science database, as of 11/2016).

Much work has been performed to investigate the mechanical and functional properties of composite hydrogels as well as their applications, and several excellent reviews are available on polymer composite hydrogels [10,11] containing various spherical inorganic nanomaterials that focused on preparation, functionalization and applications. Another review paper is dedicated to specific applications such as adsorption or drug delivery [12]. To our knowledge, no review has been made on polymer hydrogels containing carbon nanotubes and graphene sheets focusing on the composite structure–property relationships.

Fig. 1 presents the number of publications per year 2007–2016 that were obtained by searching Web of Science with keywords "clay polymer hydrogels"; "silica polymer hydrogels"; "carbon nanotubes polymer hydrogels" and "graphene network hydrogels". The publication number of polymer/silica hydrogel increases from 2007 to 2009; followed by a slight drop until 2015. Steady increments are observed for all the other composite hydrogels based on clay; carbon nanotubes and graphene; of these; polymer/graphene hydrogel shows a dramatic increase since the 1 st work published in 2010; likely due to its relatively low cost yet striking electrical conductivity and mechanical performance.

This is a critical review of the synthesis and structure–property relationship of composite hydrogels that contain carbon nanomaterials. The composite matrices are water-soluble, mainly including poly(acrylamide), poly(acrylic acid), poly(vinyl alcohol) and poly(*N*-isopropylacrylamide). While synthesis techniques are reviewed for advantages and limitations, emphasis is on the necessity to dispersing fillers at nanoscale in matrices. The mechanical performance and functional properties of the composite hydrogels are presented and discussed in relation to the morphology. Finally, the challenges and new directions in this research field are summarized.

2. Polymers for hydrogels

Polymeric hydrogels are either natural or synthetic, and they are either soluble or swollen in water. Water solubility may be

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