



# Optically triggered actuation in chitosan/reduced graphene oxide nanocomposites



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## ABSTRACT

Bio-compatible actuators which can work under optical stimulus have great future in bio-medical applications. In this work, chitosan/reduced graphene oxide (RGO) nanocomposite optical actuators were developed through a simple solvent casting technique. The photomechanical actuation of the composites is demonstrated under IR illumination. All samples exhibited contraction in length when exposed to IR light. The photomechanical stress and strain were found to increase with increasing RGO concentration. Photomechanical stress as high as 695 kPa was achieved with 4 wt.% RGO loading. Contrary to some other reported systems, the photomechanical stress decreased with the applied pre-strain. The actuation behaviour can be tuned either by altering the RGO content or applied pre-strain.

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## 1. Introduction

Polymer actuators have been widely adopted for a variety of applications due to their light weight, good processability, low cost and particularly because of their stroke, force, and efficiency being similar to that of human muscles (Ahir & Terentjev, 2005; Jiang, Kelch, & Lendlein, 2006). Depending on the type of polymer systems, polymer actuators can be triggered by an electrical, thermal, chemical or optical stimulus. Among these, optical actuators have attracted enormous interest due to their unique features like wireless actuation, remote controllability, fast recovery of the material and safety of application. These optically triggered polymeric actuators have high potential application in the field of medical devices where triggers other than heat or electricity are highly desirable.

A typical optical actuator has mainly two parts: a molecular switch unit and an energy transfer unit (Liang et al., 2009). The energy transfer unit absorbs the energy and transfers the absorbed energy to the molecular switch unit which undergoes the mechanical deformation. Basically the polymer matrix acts as the molecular switch unit and either a photoresponsive functional group or a filler acts as the energy transfer unit. Polymers with photo responsive functional groups have low photo responsive

time and mechanical property whereas in a polymer composite, by selecting an appropriate filler particle, the actuation stroke as well as the mechanical stability can be improved. Carbon nanotube (CNT) is one of the commonly used fillers in the light triggered polymer composite actuators (Ahir, Huang, & Terentjev, 2008; Lu & Panchapakesan, 2007; Yang, Setyowati, Li, Gong, & Chen, 2008; Lu & Panchapakesan, 2005; Koerner, Price, Pearce, Alexander, & Vaia, 2004; Levitsky, Kanelos, Woodbury, & Euler, 2006; Ahir & Terentjev, 2006; Ahir, Squires, Tajbakhsh, & Terentjev, 2006; Lu, Ahir, Terentjev, & Panchapakesan, 2007). However, due to the inherent bundling nature, poor dispersibility, intrinsic impurities from catalyst and amorphous carbon and high cost of CNTs, researchers are looking for improved materials to replace CNTs for practical applications.

Graphene, a monolayer of carbon atoms tightly packed into a two dimensional honeycomb lattice containing conjugated  $sp^2$  carbon atoms, has been reported for various types of actuator applications. Due to the unique properties of graphene, including high electrical, thermal and optical conductivity, graphene-polymer composites have found a steady growth in actuator research (Wang, Ko, Park, Park, & Kee, 2015; Surana, Pramod, Bhattacharya, Verma, & Mehra, 2015; Yang, Choi, Choi, Jeon, & Lee, 2012; Sen et al., 2015). Graphene, along with other members of the graphene family like graphene oxide and reduced graphene oxide, have proved their efficiency as a filler material for polymer optical actuators as well. Many researchers have investigated the optical actuation of graphene/polymer composites with various polymer systems including poly(dimethyl siloxanes), thermoplastic polyurethanes,

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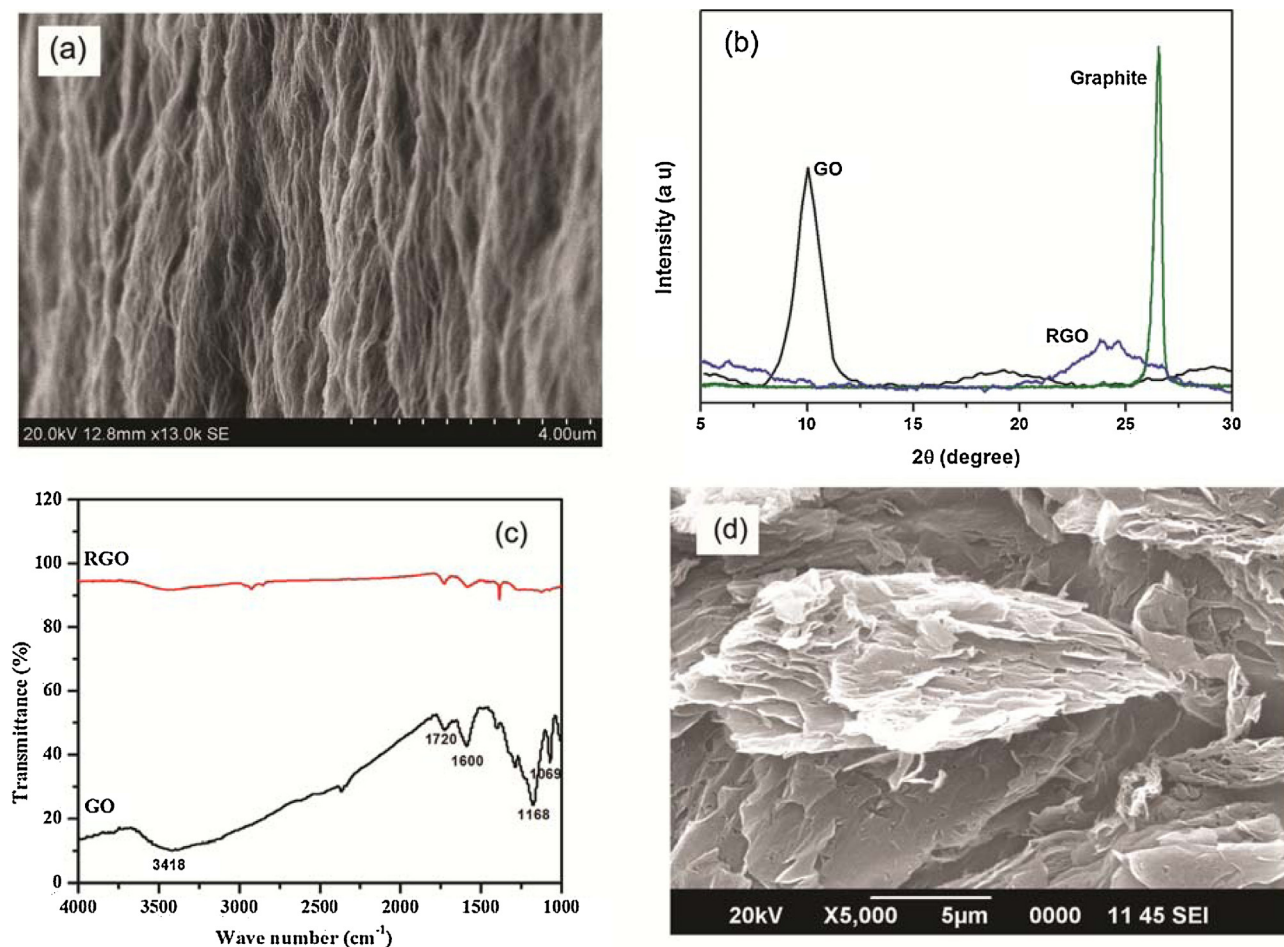


Fig. 1. (a) SEM of fractured surface of GO (b) XRD patterns of graphite, GO and RGO (c) FTIR spectra of GO and RGO (d) SEM of RGO.

liquid crystal polymers, etc. (Loomis, King, & Panchapakesan, 2012; Loomis, King, Burkhead et al., 2012; Wu et al., 2011; Seema, Rahima, & Muralidharan, 2013; Seema, Muralidharan, & Deepthi, 2013; Muralidharan & Seema, 2013). Graphene has exhibited far better efficiency than CNTs as an energy transfer unit in polymer optical actuators. For instance, Seema et al., studied the photomechanical actuation behaviour of thermally reduced graphene oxide (TRGO)/PDMS composites and found that with the same applied pre-strain and a similar loading of 1 wt.%, TRGO/PDMS composites exhibited a photomechanical stress which is 100% more than that exhibited by CNT/PDMS composite. Studies have proved that the actuation behaviour also depends on the polymer system used. For example, from our earlier studies, we have found that the optical actuation was completely reversible in graphene composites with poly(dimethyl siloxane) whereas the actuation was not completely reversible and exhibited a marching behaviour with styrene-isoprene-styrene block copolymer as the matrix (Seema, Rahima, & Muralidharan, 2013; Seema, Muralidharan, & Deepthi, 2013). Similarly, for optical actuator applications, which need very high photomechanical stress and strain, thermoplastic polyurethane matrix was found to be very effective (Muralidharan & Seema, 2013). Hence, graphene/polymer composite optical actuators can be used for many practical applications by selecting a suitable polymer matrix.

Presently, promoting sustainable development and environmental preservation is a major focus. The use of bio-compatible natural polymers is one of the important steps toward this mission. Chitosan, a copolymer of glucosamine and *N*-acetylglucosamine linked by  $\beta$ -1, 4 glycosidic bonds, is a biocompatible polymer pre-

pared from naturally abundant chitin. The biodegradable nature of chitosan makes it an environment friendly polymer and it is the second most abundant natural polysaccharide. In the last decade, chitosan composites were extensively studied as electrical/electrochemical actuators (Spinks et al., 2007; Lu & Che, 2010; Li et al., 2011; Ismail et al., 2008). However, not much study has gone into soft, biocompatible optical actuators using biopolymers like chitosan. Recently, Wu et al. reported an optical actuator based on a bi layer configuration with polyethylene as one layer and graphene/chitosan as another layer (Wu et al., 2011). They were successful in demonstrating robotic motions of a prototype actuator. However, the study was based on a bi-layer system and hence a detailed study on the optical actuation behaviour of graphene/chitosan composites alone would be still a noteworthy attempt. Moreover, in the earlier study reported by Wu et al., the effect of filler concentration was not included. Herein, we demonstrate the light triggered actuation in chitosan/reduced graphene oxide nanocomposites with excellent photomechanical stress and strain. The effect of filler concentration and applied pre-strain were also studied in detail. Due to the biocompatibility of the system, chitosan/reduced graphene oxide optical actuators can find potential applications in medical devices and the biomedical field.

## 2. Experimental

### 2.1. Synthesis of thermally reduced graphene oxide

Graphite oxide was synthesized from graphite flakes (Hind minerals, India) through a chemical route using an oxidizing mixture

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