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## **ACCEPTED MANUSCRIPT**

A Green Method for Preparing Conductive Elastomer Composites with Interconnected Graphene Network via Pickering Emulsion Templating

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#### **ABSTRACT**

The exploitation of generic strategy for preparation of conductive elastomer composites with interconnected filler network is of significant scientific and technical interests. Herein, we report a novel, green and effective strategy to prepare conductive natural rubber (NR) composites with interconnected graphene network structure. It is found that the Pickering emulsion template is effective in constructing 3D interconnected graphene oxide (GO) networks while the thermal treatment during hot pressing step is able to convert GO to conductive thermal reduced graphene (TRG). This strategy does not rely on the toxic and corrosive reducing agents and the adopted solvents in Pickering emulsions can be recycled and reused, which makes the process environmentally friendly. As a result, interconnected graphene network is well constructed within NR matrix, which significantly enhances the electrical conductivity, mechanical properties and lowers the percolation threshold of the resulted composites. Specifically, with the incorporation of 1.90 vol.% TRG, the electrical conductivity of NR composites with interconnected graphene network structure is more than 7 orders of magnitude higher than that of NR composites with uniform graphene dispersion. More intriguingly, this GO stabilized Pickering emulsion template strategy can also be used to realized the selective distribution of carbon nanotubes and assemble them into interconnected network structure, further facilitating the conductivity improvement of the resulted composites. Overall, we envision that the present work offers new insights into the design of conductive elastomer composites.

**Keywords**: Graphene; Carbon nanotube; Pickering emulsion templating; Interconnected network; Thermal reduction; Conductive elastomer composites

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