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# Monte Carlo simulations of effective electrical conductivity of graphene/poly(methyl methacrylate) nanocomposite: Landauer-Buttiker approach

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ABSTRACT

Three-dimensional Monte Carlo simulation was used to investigate the percolation phenomenon and electrical conductivity of graphene/poly(methyl methacrylate) (PMMA) nanocomposite. The electrical conductivity of this nanocomposite was investigated based on three-dimensional resistor network. By employing Landauer-Buttiker (L-B) formula for calculating tunneling resistance of a graphene-polymer-graphene junction and the intrinsic resistance of graphene sheets, transmission probabilities were estimated by scattering theory and Wentzel-Kramers-Brillouin approximation. Effects of cubic representative volume element size, tunneling distance, graphene diameter, orientation of graphene, and image potential on conductivities of graphene/PMMA nanocomposite were studied using this model. The simulation results showed that, as tunneling distance increased, volume fraction of graphenes required for creating an electrically conductive model was decreased. Also, the nanocomposite with a larger diameter of graphene had lower percolation threshold and lower conductivity. Graphene disperse in polymer with normal vector parallel to the electric field had minimum conductivity and maximum percolation threshold. Taking into account the effect of image potential increased the conductivity of graphene/PMMA and decreased percolation threshold.



## 1. Introduction

The incorporation of conductive nanofillers into nonconductive matrices such as polymeric or ceramic allows to obtain a new class of electrically conductive nanocomposites. The electrical conductivity of this class of engineering materials is strongly dependent on the volume fraction of nanofillers. Li et al. [1] indicated that effect of quantum tunneling plays a dominant role in the electrical transport of carbon nanotube–based composites. At low volume fractions, the average separation distance between nanofillers is large. Thus, the conductive paths are not formed within the nonconductive matrices. Therefore, conductivity has the values of the matrice. After increasing the volume fraction of nanofillers above the critical volume fraction (percolation threshold), the electrical conductivity of composites will increase by several orders of magnitude and, finally, will tend to be constant. This process is known as percolation process.

Electrical conductivity of polymer nanocomposites depends on many factors such as shape of nanofillers, orientation of nanofillers, contact resistance between nanofillers, temperature, etc. However, it is difficult to discern which factor does have a major role using experimental techniques. An effective way to predict the electrical conductivity of nanocomposites is through analytical and numerical studies.

Bao et al. [2,3] studied the electrical conductivity of carbon nanotube (CNT) polymer nanocomposites by Landa‘uer-Buttiker (L-B) formula. Their model predicted that the compared to multi-walled CNTs-based nanocomposites, electron tunneling resistance upon the electrical conductivity in single-walled CNTs-based nanocomposites plays a more important role. Moreover, CNT's length variability and waviness plays a more dominant role than tunneling barrier's height in percolation threshold. Gong et al. [4,5] investigated the effect of CNT deformation and CNT agglomeration on the electrical conductivity of CNT-polymer composites using Monte Carlo simulations. Takeda at al. [6] analytically and experimentally investigated the effect of nanotube geometry on electrical transport of the carbon nanotube (CNT)-based polymer composites. Hu et al. [7] predicted the piezoresistivity behavior of a CNT/polymer nanocomposite using a propose comprehensive multi-scale three-dimensional resistor network numerical model. They found that the tunneling effect and change of internal

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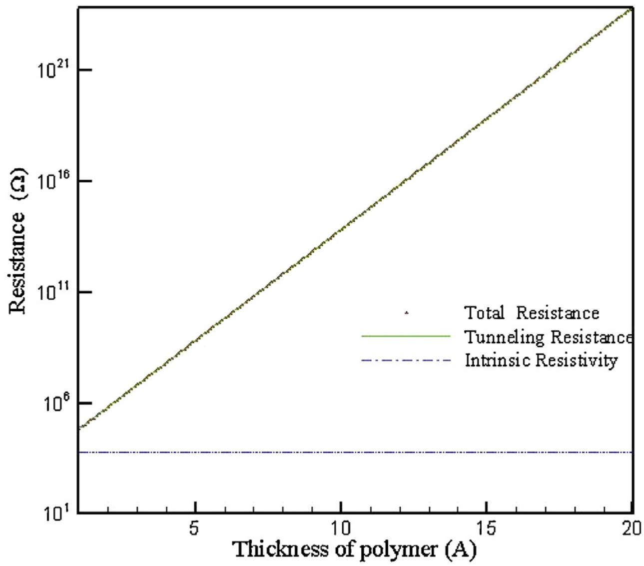


**Fig. 1.** The tunneling resistance as a function of the thickness of polymer.

conductive network were the dominant mechanisms on the piezoresistivity behavior of nanocomposite. Kuronuma et al. [8] studied the strain sensing behavior of MWCNT/polycarbonate composites under tension using analytical and experimental approaches. They figured out that the electrical resistance of the nanocomposites changed with strain.

Graphene has unique thermal [9,10], mechanical [11], optical [12], electronic [13], and transport properties [14], which contributes to its enormous potential applications as the main reinforcement filler for high performance composite materials. Since few theoretical and experimental works considered electrical conductivity of graphene based-polymer nanocomposite, further comprehensive understanding of the factors controlling conductivity will be needed in order to interpret future experiments.

Qi at al. [15] showed that, with an increase in graphene content from ~0.11 to ~1.1 vol%, the conductivity of graphene/polystyrene composites increased from ~$6.7 \times 10^{-14}$ to ~3.49 S/m. Also, they demonstrated that the percolation threshold value could be reduced by the incorporation of polylactic acid into the composite. Zhou et al. [16] reported the percolation threshold of poly(vinyl alcohol)/large-area reduced graphene oxide nanocomposites is ~0.189 wt%. Also, at the filler content of 0.7 wt%, the electrical conductivity of nanocomposites reached $6.3 \times 10^{-3}$ S/m. Park et al. [17] showed a percolation behavior in reduced graphene oxide (RGO)/polystyrene (PS) composites with the percolation threshold of _0.25 vol.% and obtained 135 S/m at 20 vol.% RGO. Hicks et al. [18] investigated the dependence of electrical transport in graphene/polymer nanocomposites on graphene sheet and device parameters by developed tunneling-percolation model using Monte Carlo simulations. Mutlay et al. [19] studied the dependence of electrical transport in graphene/poly(methyl methacrylate) (PMMA) nanocomposites on particulate geometry and temperature by a modified power law-based percolation theory. Wang and Jayatissa [20] investigated the effect of graphene size on the electrical conductivity of graphene/poly(methyl methacrylate) nanocomposite based on Monte Carlo method and percolation theory using three dimensional hard-core soft- shell rectangular prism model.

In this paper, effect of cubic representative volume element size, tunneling distance, graphene diameter, orientation of graphene, and image potential has been investigated on

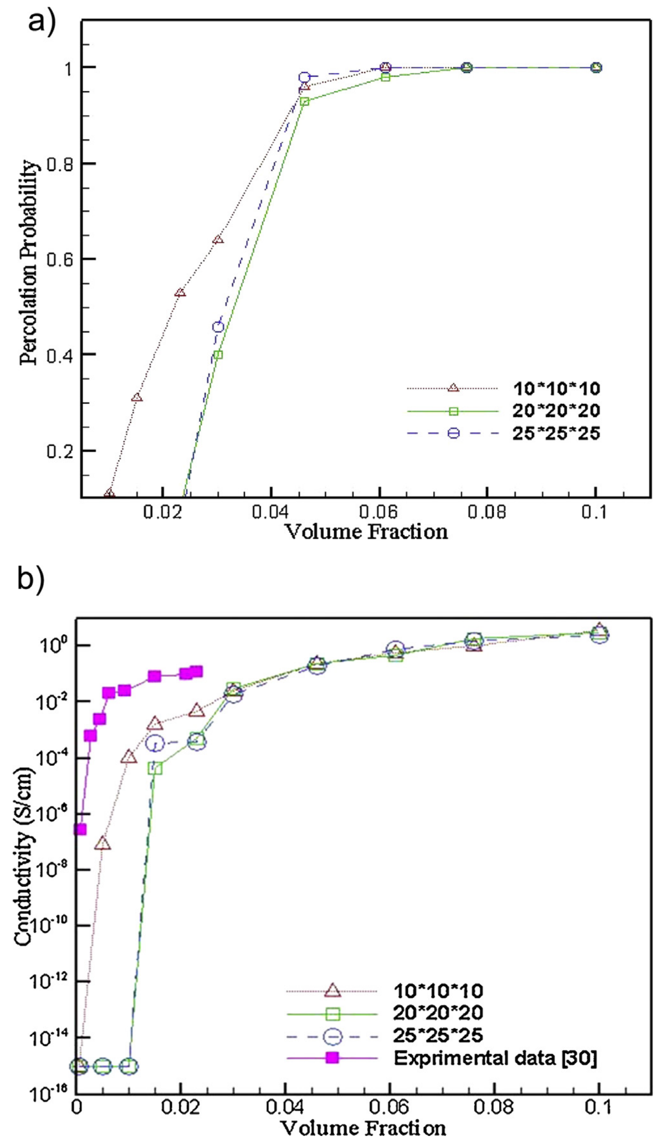


**Fig. 2.** a) Percolation probability as a function of volume fraction for the three cubes RVE. b) conductivities as a function of volume fraction for the three cubes RVE compared with the experimental data (Ref. [33]).

conductivities of graphene/PMMA nanocomposite using Landauer-Buttiker formula and three-dimensional Monte Carlo simulation. We've considered interface electrical properties with two parameter: (i)polymer as a potential barrier to electron transfer between graphene, (ii) potential image.

## 2. Model

The polymer composite was modeled as a cubic representative volume element (RVE) with edge length *L*. The graphene was modeled as hard core nanoplatelets with the thickness of t and diameter of D. The hard core nanoplatelets could not penetrate into each other. The nanoplatelets with same size were considered to be randomly scattered in RVE. Each nanoplatelets was defined by a normal vector ($n_x$, $n_y$, $n_z$) and a center point C ($X_C$,$Y_C$, $Z_C$) in [21].

$X_C = L_x$ random(1)
$Y_C = L_y$ random(2)
$Z_C = L_z$ random(3)

$$n_x = 2\mathrm{random}(4)\sqrt{1-w}$$