

Theoretical analysis of Soft Conductive Materials for Next Generation Wearable

System

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INTRODUCTION

Abstract

Purpose of the study: Recent advances in strategies for soft materials have transformed the wearable or bioelectronics from a rigid form into a soft, having advantages in terms of mechanical similarity with human tissue. Conductive nanocomposites are promising components as conductive interconnects in stretchable electronic system. This study is about optimizations of nanocomposite for enhancing its performances without degrading mechanical properties.

Methodology: First, we summarize the recent advances in metallic nanocomposites. Next, we discuss the 3-dimensional percolation theory, which is basic theoretical basis to understand the random system of nanocomposite. From this, we also briefly search important parameters having potential to change percolative connections of nanoparticles.

Main Findings: We investigated required parameters, which could affect the percolation network of conductive fillers in matrix. Dimension, shape and volume fraction of fillers are very important to realize the high conductivity of conductive composite. By calculating some parameters with theoretical formula, we analyzed the effect of shape and dimensions on performance of conductive composite.

Implications: This study can help researchers to understand the potential parameters that could affect the performances of conductive nanocomposite and analyze them in qualitative and quantitative approaches.

Novelty: The potential applications of optimized conductive nanocomposite, especially focused on wearable and bio-implantable system are discussed.

Elastic and deformable electronics have been in high demand due to their capacity for creating wearable and comfortable electronics. For instance, essential electronic devices such as sensors, (Zhou etal., 2019), actuators, (Adrian etal., 2019), energy harvesters (Minsu etal., 2019) and circuits (Park etal., 2019) have been designed in stretchable form to be used as wearable or bioimplantable systems. Thus, materials to match the qualities of these components have been widely researched, such as ultrathin metal/oxide films, and wavy/serpentine thin metals. (Fan etal., 2016). However, although these approaches do match the needs for mechanical deformations in terms of durability and sturdiness, the brittleness and rigidness of these conducting materials do not seem to be viable for mechanical human parts including organs and skins

To develop the electronics systems that minimize such mechanical mismatch between biotic and abiotic interfaces, scientists have researched nano-composites fabricated by incorporatingelastic polymers and metallic nanomaterials. Basically, stretchable nanocomposites, which are one of promising candidates for intrinsically stretchable electronics, consist of polymer matrix, functional nanoparticles and organic medium. Due to their exceptional mechanical properties (elasticity, stretchability), researchers have researched nanocomposites to improve their performances without any mechanical degradations. (Matsuhisa etal. 2017). For the importance of providing human-friendly devices that require softness comparable to human tissue, nanocomposites have great potential in providing appropriate medical technology and electronic devices in general to the world.

In this work, we highlight the most important advances in the development of stretchable and conductive nanocomposites, mainly underscoring the material synthesis and their mechanical and electrical performances. Then we investigate the parameters and theory, which can affect the electrical performance of conductive nanocomposite. In specific, we review the recently developed metallic nanocomposites having strategies to improve the conductivity. Then, from parameters we investigated, we analyze the effect of these parameters and predict what kinds of properties enhanced in nanocomposite. For examples, 3-dimensional percolation theory, which show the level of connections of conductive nanoparticles, would be addressed and we search a few kinds of parameters related to this theory to find qualitative or quantitative relationships. In addition, we would address some potential applications in wearable or bioelectronics with conductive nanocomposites enhanced by investigated parameters.

Why we need conductive nanocomposites in stretchable electronics?

Due to high Young's modulus of the conventional materials, it catalyzes mismatches at the device and skin, which prompts for inflammation to occur at the local site (Bae etal., 2013) When body parts are deformed, the external stress is applied due to modulus mismatch. Moreover, electronic devices are vulnerable to modulus mismatch, inducing



delamination of devices from skin. Recently, conductive composite, which consists of conductive nanofiller and polymer matrix, have showed good performance in stretchable and wearable electronics. By using this materials, we can minimize the modulus mismatch between device and skin, providing mechanical stability. Furthermore, unconventional properties such as electrical recovery or high electrical performance under strain, which induced by dynamic movements of conductive nanofillers in the polymer matrix are very attractive things in development of stretchable electronic devices. In the future, we could make stretchable electronic device system with high mechanical stability if highly conductive nanocomposite could be developed.

RECENT PROGRESSES IN METALLIC NANOCOMPOSITE

General metal is stiff and bulky which results in it not being compatible with soft electronics. However, metallic nanomaterials solve that issue as they can make stretchable or flexible composite materials. They have unique electrical, thermal and magnetic properties derived from their extremely small size. Like metal, metal nanomaterials have extraordinary electrical and thermal conductivities. Mixed with elastic media, the conductive nanocomposites become soft and conductive. In this soft matrix, metallic nanomaterials can make conductive pathways. These nanostructured metal materials can then be classified within their own dimensions. Examples of those dimensions are 0D, 1D, and 2D nanomaterials, which includes nanoparticles, nanowires, and nanosheets (Figure 1). 0D metal nanomaterials that are composed of gold, palladium, silver, and platinum have all been explored. However, it is still very difficult for 0D metal nanocomposites to construct the conductive percolation network. Due to this, we often add 0D metal nanocomposites to 1D and 2D metal nanocomposites to reinforce electrical conductivity without sacrificing the mechanical property of the original nanocomposites. In specific, 2D nanomaterials could make better contact between nanofillers due to having contact area while 0D and 1D nanomaterials have point contact. So, 2D nanomaterials have been used more frequently for the conductive nanocomposites. since 0D nanomaterials can strengthen the quality of the contact in the percolated network of 1D and 2D nanomaterials, they have been used together with 1D and 2D nanomaterials. This creates a hybrid type of filler materials. With this, we will show the soft conductive nanocomposites that incorporate 0D, 1D, and 2D metal nanomaterials. For example, Kim and coworkers were able to fabricate a stretchable conductor composed of gold and PU nanoparticles (Kim etal., 2013). 13 nm-sized gold nanoparticles that were citrate-stabilized (AuNPs) were blended into PU matrix via two techniques. Those two techniques were layer by layer (LBL) deposition and vacuum assisted flocculation (VAF). Both AuNPs and PU nanocomposites have the same content of filler (21.7% volume). Five films for both techniques were used to analyze mechanical and electrical properties of the nanocomposite. Also, Kim et al. were able to demonstrate elongation of conductive composite which broke at 1780% by using MIBK, stretchable rubber, and Ag flakes. They showed its feasible applications in wearable sensor technology. In other paper, researcher showed that degraded electrical performance under high strain could be recovered with dynamic arrangements of nanofillers in nanocomposite (Kim etal., 2019).

3-Dimensional percolation theory

Percolation theory in nanocomposite deals with the behavior of some geometrical and physical properties related to connectivity in a matrix system. In classical percolation theory, researchers follow these properties assuming a well-defined-geometrical criterion for the inter-element bonding, and thus the minimal concentration of bonds that yields connected network is defined as the percolation threshold (<u>Balberg etal., 2004</u>). Such percolation systems are manifested in various porous media and other complex polymer matrix, and they are characterized in the continuum by percolation thresholds that are given by a critical fractional volume of nanomaterials, *Vc*. The value of *Vc* can be in the whole range between almost 0 and the close packing values of the corresponding nanomaterials (Figure 2). The percolation theory was applied to explain electrical conductive behavior of composites. Near the percolation threshold, the electrical conductivity of composites follows a power-law relationship where σ is the electrical conductivity of sis a conductivity exponent. Because Equation 1 cannot take into account either of particle shape, orientation, polymer–particle interaction or particle dispersion, the value of s is not a constant, but varies with particular composite systems. s and V_c have to be determined by curve fitting of experiment results.

$$\sigma = \sigma_0 (V_{\rm f} - V_{\rm c})^s$$
 (Equation 1)

Conductivity and critical volume fraction can be defined as intrinsic electrical properties of nanomaterial. Beyond this equation, there are many geometrical and physical parameters which can affect s value including shape of nanomaterials, density of polymer chain. We will consider these parameters in next section.

Shape of nanomaterials

To make conductive nanocomposite, we can use many kinds of conductive nanomaterials having different geometries such as 0D, 1D, 2D, and 3D as we mentioned above. To investigate the effect of geometries, 2D and 3D randomly distributed platelets are considered and the influence of geometric shape of platelet is specifically evaluated in this section. To obtain the relationship between volume fraction of nanomaterials and other parameters, distance between adjacent conductive particles is calculated. It is assumed that the particles are homogeneously distributed within the matrix and fixed in unit cubic of polymer matrix. Two cases are considered: i) 0D spherical nanoparticles and ii) 2D square-like nanoplate. If the particle has a shape of sphere, as shown in Figure 3a, the length of the unit cubic of polymer matrix, L, is presented by equation 2.



 $L = D + I = (\pi D^3/6V_f)^{1/3}$, $V_f = V_{nano}/L^3$ (Equation 2)

where D and V_f are the diameter and the volume fraction of particle, I is the distance between particle. Rearranging equation 2 gives below (Equation 3).

$$V_{f} = \pi D^{3}/6(D+I)^{3}$$
 (Equation 3)

The critical volume fraction of spherical particle in the composite is a function of diameter of particle and distance between particles. From this, we can calculate the required volume fraction of spherical nanoparticles to form conductive connections.

In case of 2D square-like nanoplate (Figure 3b), we assume that there is no angle variance. From this, we can also express the V_f as a function of D and I as below shown in Equation 4 and 5.

$$L = D + I = (D^2 t / V_f)^{1/3}, V_f = V_{nano} / L^3$$
 (Equation 4)

$$V_f = D^2 t/6(D+I)^3$$
 (Equation 5)

From these calculations, we can expect the minimum amount of conductive nanofiller for fabricating conductive nanocomposite.

POTENTIAL APPLICATIONS OF CONDUCTIVE NANOCOMPOSITE

There have been many researches to minimize the modulus mismatch between biological tissue and bioelectronics for diagnosis of disease. To improve the accuracy of biological sensors, we have to make electronic components attached on tissue with high conformability and durability. Due to dynamic movement of organs in our body, the electronic device is fabricated by soft materials to relax the external stress. Various kinds of conductive nanocomposites have been researched for a long time to fulfill these requirements and many progresses have been shown in bioelectronics (Sim etal., 2020). Moreover, conductive nanocomposites are widely used in wearable electronics to connect the chips including physiological sensor, wireless antenna and logic components (Lim etal., 2021). From these soft connections, all electronic chips could be well operated under harsh stress. Finally, soft electronics also need these kinds of soft conductive nanocomposite. Recently many kinds of prosthetic hand have been fabricated with metal or steel that could be harm to human. By replacing these materials with soft nanocomposite, we can form a human-friendly soft robotic system for patient and people involved in human-machine interfaces.

CONCLUSION

In this research, we consider the promising technologies regarding metallic nanocomposite. First, we review the previous works, which showed the electrical performance under strain and its mechanisms. Then we research the fundamental theory that could affect the performance of conductive nanocomposite. Percolation theory is well known theory to understand and predict the network of composites. From this theory and mathematical approach, we can expect the critical volume fraction of nano fillers in a few cases such as 0D and 2D. Finally, we address the potential applications with conductive nanocomposites.

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AUTHOR'S CONTRIBUTION

All the author(s) have contributed equally in the preparation of the manuscript.

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FIGURES



Figure 1: Schematic showing the nanocomposite with different shapes of nano fillers



Figure 2: Conceptual graph showing percolation threshold, termed V_c



Figure 3: Schematic showing model of composite in different shape of nanomaterials