

Heat-Sink Solution Through Artificial Nanodielectrics for LED Lighting Application

N. Badi^{1*} and R. Mekala²

¹Physics Department, Center for Advanced Materials, University of Houston, Houston, TX, USA

²Electrical & Computer Engineering, University of Houston, Houston, TX, USA

* Corresponding author: nbadi@uh.edu

Abstract: Thermally conducting but electrically insulating materials are needed for heat-sink LED lighting applications. We report on a cost effective and innovative method based on creating core-shell nanoparticles in polymer with aluminum (Al) nanoparticles as the high thermal conductivity core, and ultrathin aluminum oxide (Al_2O_3) as capping shell for electrical insulation. The solid oxide shell around the Al core prevents agglomeration of Al nanoparticles. A coupled COMSOL heat transfer and AC/DC modules were used in the simulation. The mean effective thermal conductivity seems to exceed the value of 100W/m.K with an electrical permittivity of 52 at 12% Al- Al_2O_3 core-shell nanoparticles loading in a polyvinylidene fluoride polymer nanocomposite (PVDF). These results are considered outstanding but they need to be validated experimentally.

Keywords: Heat-sink LED lighting, Nanodielectrics, Core-shell fillers, Metal nanoparticles, Effective medium theory

1. Introduction

Light Emitting Diodes (LEDs) are likely to replace other technologies such as incandescent and fluorescent bulbs in signaling, solid state lighting and vehicle headlights because they save energy and extend the light's lifetime. Thermal management of LEDs is a crucial area of research and development [1]. High performance LEDs requires superior thermal management systems. Heat sink mounts that dissipate heat generated by these high power LEDs are of critical importance for this purpose. The advent of more sophisticated applications of heat-sink materials has led to tremendous interest in alternative methods that can provide access to thermally and electrically stable materials. Composites made with epoxies and polymers mixed with various additives have been developed and are available in the market [2-6].

All these materials lack either in one or more than one parameters which are important for optimal properties.

2. Approach

Thermally conducting but electrically insulating materials are needed for heat-sink LED lighting applications. Metals are thermally and electrically conductive, so they cannot be used. All polymers are electrically and thermally insulated. Thus, polymers alone cannot be used for this application. However, polymers can be modified with mixing or compounding them with shells-isolated (capped) metal nanoparticles which are thermally conductive and at the same time electrically insulated (Figure 1). Such nanoparticles with ultrathin shells allow the surface of a particle to be altered while the particle maintains its bulk properties.

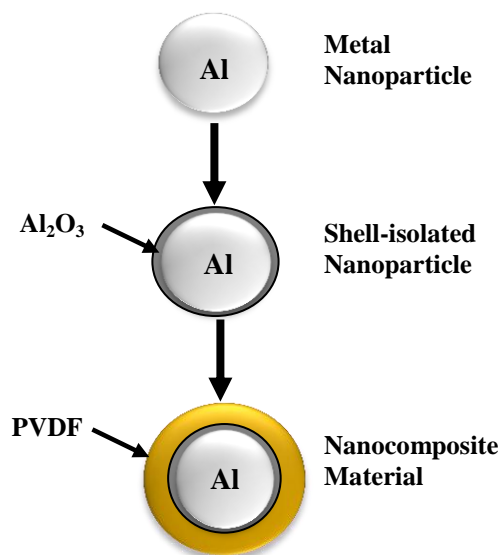


Figure 1: Representative scheme for oxide shell-isolated metal nanoparticle in PVDF polymer.

Al nanoparticles are highly desirable as a thermal conduction medium to make high thermal conductivity nanocomposite materials with controlled dielectric strength for LEDs lighting applications. One difficulty with Al nanoparticles is the relative inertness of their surfaces. This inertness limits the loading of Al nanoparticles in the polymer composite. Higher loadings are needed as the heat-sink industry develops faster and denser integrated modules that produce more heat. This can be achieved by coating Al nanoparticles with a more suitable inorganic film like Al₂O₃ film. The solid oxide shell around the Al core prevents agglomeration of Al nanoparticles. This physical barrier overcomes the internal charging creation which in turn improves the dielectric strength of the nanocomposite. Furthermore, the shell-isolated structure enhances the thermal and chemical stability of the nanoparticles, improves solubility and reactivity as well as allows conjugation of other molecules to these particles.

3. FEM simulation

With advancements in computer technology, simulating complex percolative systems like nanocomposites has become a reality. Using the finite element method available in COMSOL Multiphysics, effective properties of the composites can be calculated. Primary goal was to calculate both the effective heat thermal conductivity and electrical permittivity of the composite medium formed by PVDF and Al-Al₂O₃ nanofillers. Effective medium theory (EMT) was used to calculate the effective electric permittivity of the nanocomposite [7]. The mean effective thermal conductivity of the nanocomposite was estimated by integration of thermal heat flowing across the constituent components of the nanocomposite.

3.1 Thermal conductivity

The heat-transfer equation and boundary condition (included in the model using the General Heat Transfer application mode) used to describe the effective thermal conductivity are:

$$\nabla \cdot (-k\nabla T) = Q \quad (1)$$

$$-\mathbf{n} \cdot (-k\nabla T) = h(T_{inf} - T) \quad (2)$$

Where k is the thermal conductivity (W/(m·K)), Q is the heating power per unit volume of the LED, \mathbf{n} is the normal vector of the boundary, and h is the heat transfer film coefficient (W/(m²·K)), and T_{inf} is the temperature (K) of the surrounding medium.

3.2 Effective Medium Theories

Effective medium theories (EMTs) are used to calculate effective properties of a medium with inclusions. EMTs and other mean-field like theories are physical models based on properties of individual components and their fractions in the composite [8]. Generally, properties calculated using EMTs are dielectric constant and conductivity. There are many EMTs, and each theory is more or less accurate under different conditions.

Most popular EMTs [7, 9] are those of Maxwell

$$\varepsilon_{eff} = \varepsilon_h + 3f \frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h} \varepsilon_h \quad (3)$$

Maxwell – Garnett model

$$\varepsilon_{eff} = \varepsilon_h \left[\frac{1 + 2f \left(\frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h} \right)}{1 - f \left(\frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h} \right)} \right] \quad (4)$$

Looyenga model

$$\varepsilon_{eff} = \left[\left(\varepsilon_i^{\frac{1}{A}} - \varepsilon_h^{\frac{1}{A}} \right) f + \varepsilon_h^{\frac{1}{A}} \right]^A \quad (5)$$

Where ε_{eff} is the effective dielectric constant of the medium, f is the volume fraction of the filler, ε_i is dielectric constant of the Al filler, ε_h is dielectric constant of the host PVDF matrix and A is a depolarization factor, which depends on the shape of inclusions. Value of A is 2 for disk fillers and 3 for spherical fillers.

3.3 Percolation

Percolation was originally studied as a mathematical subject but its broadest applications are found in the field of material research [10]. To model disordered system, one of the easiest mechanisms is percolation as it has very little statistical dependence. It is easy to realize even for most complex systems and its outcomes are not unrealistic for qualitative predictions of random composites [11]. Concept

of percolation reaches its significance when the minor phase of the composite (fillers) loading reaches a critical value when substantial changes takes place in the physical and electrical properties of the system, sometimes in order of more than ten folds. This critical fraction of filler is termed as percolation threshold, f_c . The abrupt changes in the properties of the system are particularly predominant if the constituent components of the composite have a large difference in their properties. Al filler and PVDF matrix considered in this work have highly varying electrical properties. Al has a high electrical conductivity of $3.5 \cdot 10^7 \text{ S}\cdot\text{m}^{-1}$ while PVDF has an extreme electrical conductivity of $0.2 \cdot 10^{-14} \text{ S}\cdot\text{m}^{-1}$.

Simple power law expression describes the significant changes in properties of the system near the percolation threshold.

$$\frac{K}{K_m} = |f - f_c|^{-s} \quad (12)$$

Where K is the dielectric constant of the composite, K_m is the dielectric constant of the matrix (PVDF), f_c is percolation threshold, f is the filler volume fraction and s is an exponent of value about 1.

3.4 Use of COMSOL Multiphysics

In the COMSOL Multiphysics software, the general heat transfer module is selected and both steady and transient analyses were applied to calculate the thermal conductivity of the nanocomposite. Effective Medium Theories (EMT) of Maxwell-Garnett and Looyenga models were employed to estimate the effective electrical permittivity using In-plane electric currents within the coupled AC/DC module. This model was selected because it allows a frequency sweep to be conducted for different loading fractions of nanofiller from 0 to 1. The nanofiller we used in the simulation is made of Al-core and Al_2O_3 shell and has a total diameter of 45nm, with the core as 35nm diameter.

In order to simulate the effective properties of the nanocomposite, electrical conductivity of the materials and their dielectric constants were used as subdomain conditions.

We set different values for each subdomain using material properties, sources, and sinks that define

the physics in the subdomain and which are available in the COMSOL Modules we used.

The boundary conditions define the interface between the model and its surroundings. In calculating the effective thermal conductivity of the nanocomposite, we set one of the matrix boundaries to a constant heat source at a defined T_{inf} while all other boundaries were thermally insulated. As for the electrical permittivity, we set one of the boundaries to a port as the input electrical field while its opposite boundary was maintained at ground. All other boundaries were electrically insulated. This makes the geometry analogous to a parallel-plate capacitor with an electrical field applied across its plates.

4. Results

Figure 2 shows a geometry setup of 3D model nanodielectric containing core-shell configuration made of 4 spherical nanoparticles with core diameter of 35nm and shell thickness of 10nm. The spheres are at equal spacing with each other in an enclosing cube of side length of 250 nm. This corresponds to a filler volume fractions of about $f = 0.12$.

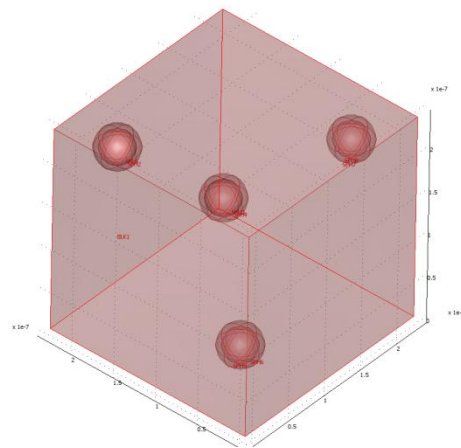


Figure 2: 3D model nanodielectric with filler fraction loading of $f = 0.12$

Simulation results with 12% Al- Al_2O_3 core-shell nanoparticles loading in a Polyvinylidene fluoride (PVDF) indicates a minimal dielectric loss of the embedded host polymer and at the same time provides a powerful approach for thermal management application for LEDs lighting application. The mean effective thermal

conductivity seems to exceed the value of 100 W/m.K (Figure 3) with an electrical permittivity K of 52 at the same fraction of loading. Figure 4 shows a transient of heat conduction analysis at a period $t = 10^{-15}$ s. The heat flows through the polymer and is conducted uniformly throughout the metal when subjected to a heat source at $T_{inf} = 230$ °C.

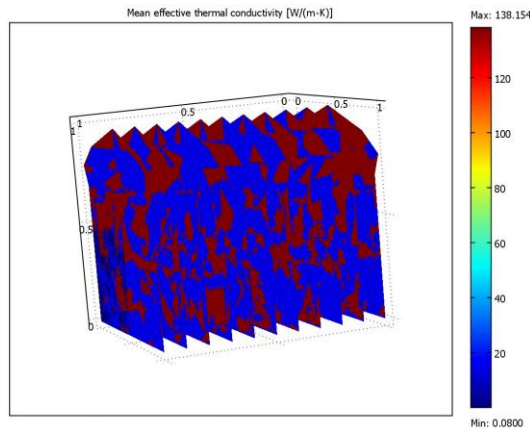


Figure 3. Mean effective thermal conductivity of Al-Al₂O₃/PVDF artificial nanocomposite

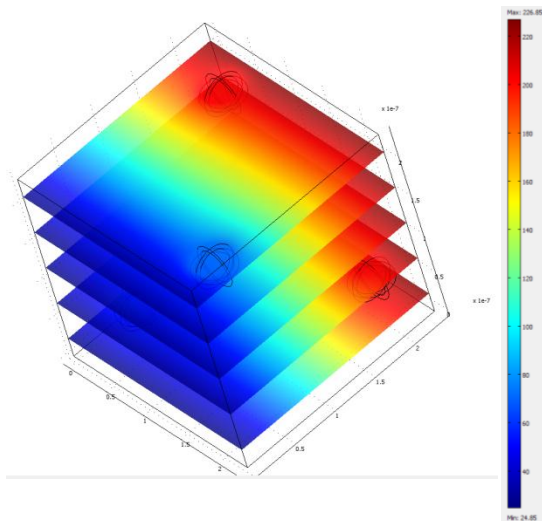


Figure 4. Transient heat conduction analysis of the nanocomposite at a period $t = 10^{-15}$ s and $T_{inf} = 230$ °C.

Figure 5 shows a gradual increase in electrical permittivity K with the increase in loading of nanofillers. As the loading approaches the critical value of the percolation threshold which is about 21%, there is a significant increase in K values of the composite up to 2650. If loading of nanofillers is increased beyond percolation threshold, composite becomes conductive in nature. This leads to a rapid fall in the dielectric constant.

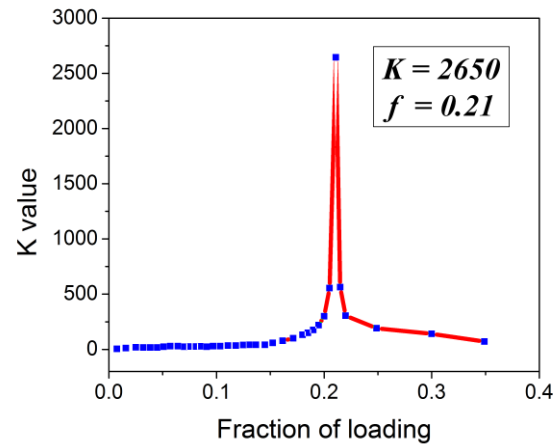


Figure 5. Predicted K value of 2650 with Al-Al₂O₃-PVDF nanocomposite at a volume loading of 21%

5. Conclusion

COMSOL heat transfer and AC/DC modules were used in the simulation of heat-sink solution for LED lighting application. The mean effective thermal conductivity seems to exceed the value of 100W/m.K with an electrical permittivity of 52 at 12% Al-Al₂O₃ core-shell nanoparticles loading in a polyvinylidene fluoride polymer nanocomposite (PVDF). These results are considered outstanding but they need to be validated experimentally.

It is anticipated that proper loadings of engineered core-shells in selected high dielectric strength thermoplastic polymer will provide high thermal conductivity, enhance the dielectric properties of polymer by mitigating the internal charge creation within polymer host while maintaining electrical insulation to dissipate the heat with ultrafast response.

6. References

1. M. Arik, A. Setlur, S. Weaver, D. Haitko, and J. Petroski, Chip to System Levels Thermal Needs and Alternative Thermal Technologies for High Brightness LEDs, *Journal of Electronic Packaging* 129, 328 (2007).
2. M. Pietralla, High thermal conductivity of polymers: Possibility or dream?, *Journal of Computer-Aided Materials Design* 3, 273 (1996).
3. Ye.P. Mamunya, Electrical and thermal conductivity of polymers filled with metal powders, *European Polymer Journal* 38, 1887 (2002).
4. Jacob M. Wernik and Shaker A. Meguid, Recent Developments in Multifunctional Nanocomposites Using Carbon Nanotubes, *Applied Mechanics Reviews* 63, 050801 (2010).
5. L H Liang *et al.*, Thermal conductivity of composites with nanoscale inclusions and size-dependent percolation, *J. Phys.: Condens. Matter* 20, 365201 (2008).
6. Weixue Tian and Ronggui Yang, Phonon Transport and Thermal Conductivity Percolation in Random Nanoparticle Composites, *Tech Science Press CMES* 24, 123 (2008).
7. V. Myrochnychenko and C. Brosseau, “Finite-element method for calculation of the effective permittivity of random inhomogeneous media”, *Physical Review E* 71, 016701 (2005).
8. Tuck C. Choy, “Effective medium theory – principles and applications”, *International series of monographs on Physics*, Oxford science publications, 1999.
9. William M. Merrill, et al, “Effective Medium Theories for Artificial Materials Composed of Multiple Sizes of Spherical Inclusions in a Host Continuum”, *IEEE Transactions on antennas and propagation*, vol. 47, no. 1, January 1999.
10. C. W. Nan, Y. Shen and Jing Ma, “Physical properties of composites near percolation”, *Annu. Rev. Mater. Res.* 40, 3.1–3.21 (2010).
11. Geoffrey Grimmett, “Percolation”, Springer link, 1991.

7. Acknowledgments

This material is based upon work supported by the Texas State Fund to the University of Houston Center for Advanced Materials (CAM).