Thesis Changes Log

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PhD Program: Materials Science and Engineering

Title of Thesis: Material design and optimization of thermal management materials based on boron nitride, graphene, and carbon nanotubes polymer nanocomposites

Supervisor: Prof. Sergey Abaimov

The thesis document includes the following changes in answer to the external review process.

I am really grateful to all the Jury members who spent their time to my thesis. The answers to the comments are the following:

Name of the Reviewer: Igor Shishkovsky

1. The author has conducted a comprehensive investigation into the thermal and electrical properties of synthesized polymer-filled composites. However, there are certain aspects related to heat transfer at the interface that have not been addressed. Heat transfer at the interface, particularly in cases of different media such as solid-liquid and solid-gas interfaces, plays a crucial role in determining the efficiency of heat removal from the heated object.

   The required topic related to interface is added to the updated thesis. Please see page 30, section 2.2.4.

2. When discussing the measurement of the wetting angle in paragraph 3.43 or 4.4.2.1, the author has not considered the impact of temperature changes on the heat sink. It is important to note that the coefficient of surface tension, and consequently the value of the wetting angle, is influenced by temperature variations.

   Coefficient of surface tension has indeed a significant role in changing the wetting angle. Unfortunately, we didn't check this phenomenon when observing the Water contact angle with PVA and its nanocomposites films or 3D bulk structure. We will surely consider this in our future research.

3. In paragraph 3.46, the diffraction peaks are mentioned as (002), (100), and (004), but there is a need for further clarification and specification. In the provided Appendix 1, the author has included explanations; however, it would be beneficial to associate each diffraction peak with specific materials. As different modifications of the BN phase are recognized, providing separate diffractograms for PVA and BN would help elucidate the situation and enhance clarity.

   The XRD graph is replotted with independent diffractograms for PVA and BN. The explanations are given. Please see section 3.4.6, page 65.

4. Contents of the Second Chapter (Page 12): It appears that Paragraph 2.3 is duplicated within this chapter. –

   Corrected

5. Paragraph 2.1.1: It is not immediately clear in this paragraph about the distinctions between thermal 'paste', 'grease', and 'pad'. –

   Thermal paste is a semi-liquid material, usually a mixture of thermally conductive particles (like metal oxides) suspended in a silicone-based or other non- electrically conductive carrier fluid. Thermal paste is applied in a thin layer between the heat source and the heat sink to fill microscopic imperfections and enhance thermal conductivity. However, like thermal paste, thermal grease typically contains thermally conductive particles in a non-conductive carrier fluid and they have a very high viscosity. Whereas, thermal pads are solid-state materials that come in the form of soft, compressible pads or sheets. They are usually made of silicone or other polymers impregnated with thermally conductive fillers. These are placed between the heat source and the heat sink. They do not require spreading or application like thermal paste or grease. Instead, their soft, compressible nature allows them to conform to the surfaces when pressure is applied.

6. Caption in Figure 3: The significance of blue and red particles in Figure 3 is not explained in the caption. –

   Blue and red particles depict the representation of polymeric atoms. In discontinuous networks, a large amount of phonon scattering would occur at the filler/matrix interface because of the acoustic mismatch occurring in different atoms (High phonon scattering between green and blue, red atoms as shown in the Figure 5b in the updated thesis). Substantially, the optimization of the filler structure (including the size and aspect ratio) and the filler dispersion can enhance the thermal conductivity by arranging the filler to form a thermally conductive network in the polymer matrix.

7. Figure 5: The figures labeled as 'a)' and 'b)' are missing in Figure 5.

   Corrected.

Name of the Reviewer: Alexander Korsunsky

The suggestions by the reviewer are noted and a sufficient amount of literature is added in the updated thesis. Please see section 2.2.1 and 2.2.2. Page numbers 24 to 28. The abstract is also reduced.

Name of the Reviewer: Albert Nasibulin

1. Abstract: Guidelines dictate that the abstract size is limited to 400 words. Currently, the abstract is over 700 words long.

   Abstract is cut to short.

2. The list of figures and tables is not necessary and can be easily cut off without harming the quality of the thesis. This is old style, I would call atavism.

   List of figures and tables are removed from the updated thesis.

3. Section 1.2: "To achieve maximum improvement in the thermal conductivity of TMMs or TIMs compared to the traditional TMMs currently employed in industry with TC of ~ 4W/mK by investigating the effect of low (≤ 10) to optimum filler loading concentration within a polymer matrix." What is 10? 10% by weight? By volume?

Filler loading concentration is by weight. It is edited in the updated thesis.

4. Section 2.2: It may be beneficial to describe the difference between thermoset and thermoplastic polymers. The thesis does a good job in explaining their usages and applications, but misses to explain their difference.
   The difference is explained and the thesis is edited. Please see section 2.2, Page 22 and 23.

5. Section 3.3 – SEM details are missing.

   Edited.

6. Sections 1.2 and 2.1 should start with a short description. For instance, what section 1.2 contain: objectives or scope? The same comment for "2.11" and please check the numbering.

   Edited

7. Abbreviations must be given in an alphabetic order. Descriptions for Tables should be placed above the tables, but not below.

   Edited

8. Chapter 6, which is conclusions, summarized the thesis and connects it well, it would be nice to see this interconnectivity logic in the final paragraphs before switching chapters.

   As per your suggestion, work is done by connecting chapters with each other please see sections 2.5 and 3.6.

Name of the Reviewer: Jun Zhao

Language of the thesis is improved as per suggestion. Please see the updated thesis.

Name of the Reviewer: Qiang Liu

1. In chapter 3, the author interprets the effects of BN content on the overall TC (Fig. 7) in the view of homogenous dispersion and nano-level structure. It may be more convincing if the author can give some SEM pictures corresponding to different BN contents, such as those in Fig. 6. I think it can better help readers to under the interpretation.
   The author believes that the ratio of 2:1 is an optimum ratio by comparing three different ratios. Has the author tried the ratio smaller than 2:1, e.g. 1:1?
   The same questions for the investigations of electrical resistivity.

   As per your kind suggestion, more, SEM images are added in the section 3.4.4 with Fig. 7. In the updated thesis.
   We didn't try the ratio of 1:1 because normally the literature study on BN/PVA aerogel composites showed the highest TC for BN/PVA aerogel with the loading weight fraction of BN as ~ 70 % in a PVA based segregated structure. But the nevertheless, future work on this 1:1 ratio could be done for better understanding of possible connectivity and thermally conductive paths of BN in PVA segregated structures.

   Whereas, the investigation regarding electrical resistivity is concerned, I assume based on literature review that the system will not make a significant change in the electrical insulation of BN/PVA composites as the BN being electrical insulator and having a high electron band gap will increase the overall electrical resistivity of the system as shown in the other different ratios of BN/PVA, in Figure.8 respectively but still we will consider your suggestion for our future research work.

2. A minor question: the four subpictures in Fig. 5 corresponds to the BN/PVA ratio of 0:1, 9:3, 9:1 and 2:1, respectively. But the sequence of the four ratios are different in Figs. 7 and 8.

   The sequence of four ratios is edited in Fig. 5 which are in coherent with the sequence with Fig. 7 and 8.

3. In chapter 5, for the two types of MWCNTs, the size of CNT agglomerates seems to has no significant on the electrical and thermal conductivity of nanocomposite according to the experimental results in this thesis, can we have a such a conclusion? Or are there some other factors leading to such experimental results, e.g. the poor impregnation of epoxy into CNT agglomerates? Especially, as shown in Fig.4, increasing the content of MWCNTs has very limited effects on TC of nanocomposites. I think the author would add some interpretation to address the effects of the dispersion and contents of MW1 and MW2 which have been obtained in this work. As there are a great number of report in literatures, it is better to give some comparisons between the present results with those in literatures, especially for the effects of MWCNT agglomerates, which could help the author to better interpret the present experimental results.

   In the case of Single-Walled Carbon Nanotubes (SWCNTs), a discernible percolation network emerges within the epoxy resin system, comprising interconnected SWCNTs and SWCNT bundles with homogenous dispersion. This network formation signifies a notable enhancement in thermal conductivity. However, in the case of Multi-Walled Carbon Nanotubes (MWCNTs), we observe the presence of ovular agglomerates, and huge vacant regions of epoxy resin as depicted in the SEM images in Figure 2. These images clearly reveal the impregnation of epoxy resin into the spaces between MWCNTs. Unfortunately, the percolation networks formed by MWCNTs in other regions are not sufficiently well-structured to yield a significant increase in thermal conductivity like we see in SWCNTs. The discussions are added in the section 5.41.