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Thesis Changes Log

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PhD Program: Materials Science and Engineering

Title of Thesis: CARBON NANOTUBE FIBERS AS EMBEDDED ELECTRODES FOR THE DUAL-STAGE MONITORING OF MULTI-FUNCTIONAL CARBON NANOTUBE NANOCOMPOSITES

Supervisor: Prof. Albert G. Nasibulin

Co-supervisor: Asst. Prof. Dmitry V. Krasnikov

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

Firstly, I would like to take the opportunity to thank the Jury Members of the Reviewing Committee. I would like to acknowledge and appreciate the time and effort that they have spent on my dissertation and for the constructive comments and suggestions provided for improving it. The responses to the comments and suggestions provided by each Jury Member have been provided below:

Comments from Professor Oleg Tolochko:

1. ***It is not clear how the concentration of nanotubes in the composites for study was selected. In general, composites with nanotube concentrations of up to 2% were prepared, but only two concentrations of CNTs (0.25% and 0.75%) were used for the study, which were used to determine the sensitivity of CNTFs to the properties and concentration of CNTs.***

Response:

Professor Tolochko has raised a valid comment regarding the selection of weight percentages for nanocomposite investigation. Several factors were considered when choosing the amount of CNT addition, which are as follows:

1. Background studies conducted within the duration of the PhD examined weight percentages of a higher values (0.5, 1.0 and 2.0 % weight) [1,2]. For the multi-walled CNT (MWCNT) nanocomposites, the lowest weight percentage provide electrical resistivity values on the border of electrically conductive. For the single-walled CNT (SWCNT) nanocomposites, resistivity values were much below values reported in literature. For the studies focused on in the dissertation, weight percentages not studied previously were chosen to obtain new data sets and expand on values already known (below previously studied values and in between for 0.25% and 0.75%, respectively).
2. Since this was a proof of concept and no relevant literature was found on interaction of CNTs and carbon nanotube fibers (CNTFs), weight percentages which would provide different properties were selected. For MWCNT nanocomposites, 0.25% by weight addition provided electrical properties on the border of electrically insulative and conductive, whereas 0.75% provided relatively conductive values equal to or higher than what are seen in literature [3–5]. This allowed understanding of how the CNTFs detected MWCNTs, their percolation network-based properties at different concentrations, and if the detection behaviour differed from SWCNT dispersions. The SWCNT nanocomposites were chosen at the same weight percentages because they provided the opportunity for detection of

SWCNTs, better developed percolation networks with different electrical properties and provided a weight percentage-based comparison.

3. A practical aspect was needed to be accounted for. Although preliminary investigation in this dissertation conducted experimentation and analysis until 2% by weight [6], it was seen that weight percentages above 1% were extremely difficult to process due to their pronounced viscosity. In order for the CNT nanocomposites and CNTFs to interact, a degree of material flow was needed during the molding process. This contributed to selecting the lower weight percentage values.

The comment from Professor Tolochko has pointed out that these factors were not clearly stated in the original dissertation work. Appropriate changes to make the work clearer and more understandable have been made in chapter 4.

2. ***In general, for each type of filler there is an optimal value of its concentration, at which the maximum value of the strain sensitivity coefficient is achieved. Increasing the filler concentration may lead to two different effects. Firstly, the range of deformations in which the composite retains its conductive properties increases, which should lead to a decrease in the value of GF, and secondly, the value of the resistance of the composite in the absence of deformations decreases, which should lead to its increase. The lower the filler concentration in the matrix, the less stretching the sample begins to change its resistance. A significant increase in filler concentration leads to a decrease in the sensitivity of composites to tension: in order to noticeably change the resistance, such samples had to be stretched to a greater length. This effect of filler concentration on tensile sensitivity can be explained by the fact that when stretched, the distance between particles increases, which, from the point of view of conductivity values, is equivalent to a decrease in filler concentration. In addition, stretching the composite leads to a change in the value of the critical conductivity index t, which indicates a change in the dimension of the structure, and hence the value of the percolation threshold, because the percolation threshold generally depends on both the dimension of the structure and the ratio of the sizes of conducting clusters and non-conducting sections. So, some important properties of composites should be studied for more details, such as percolation limit, localization radius of the wave function (jump length), features of the three-dimensional network presumably formed by nanotubes, etc.***

Response:

Professor Tolochko has provided an insightful and useful set of comments regarding the topic of the thesis and relevant mechanisms of property change. The Professor is completely correct in saying that each type of filler has a specific optimal value of concentration for properties such as electrical conductivity [7,8], strain sensitivity [9] or mechanical performance [10]. Increasing the filler concentration is also well reported to cause a decrease in the gauge factor of the nanocomposites, as an increase in the concentration leads to higher electrical conductivity associated with a stronger percolation network [11]. The same is valid for vice versa, where lower concentrations lead to weaker formed networks which consist of fewer nanoparticles per unit volume. Nanocomposites with higher concentrations of nanoparticles forming a percolation network react to the same amount of strain or force with a subdued piezoresistive response as compared to nanocomposites with a lower amount since it is more difficult to disrupt the connections of the network [12]. To obtain the same amount of piezoresistive response to strain, the Professor has rightly stated that a higher strain value is needed.
Here, I would like to point to one of the objectives of the dissertation. As investigating the interaction of both the CNTs and CNTFs was the main purpose of the study, the amounts of addition of the different types of CNTs were selected in such a way as to produce nanocomposites which were electrically conductive, yet showed a measurable degree of piezoresistive response. Although studying the nanocomposites at or near the percolation threshold for each type would have led to higher piezoresistive response, the study used values which were above the percolation threshold since the detection ability of the CNTFs was unknown. Hence, relatively "safe" values above the percolation threshold were chosen. It should be noted that for the SWCNT nanocomposites, as part of one of the publications of this thesis [6], the percolation threshold was investigated, and found to be φ_c (wt.%) = ~0.001, with a t value of ~1.92. The MWCNT nanocomposites have been well investigated previously with similar processing procedures, showing a percolation threshold of φ_c (wt.%) = ~0.02 [4,13]. The values of electrical conductivity achieved by the MWCNT nanocomposites match values in this study at the selected weight percentages. The difference between the two types of

nanocomposites and their critical parameters is associated with their aspect ratios, which has been very kindly pointed out by Professor Tolochko. Relevant changes to the text concerning these reasons behind the choice of investigated weight percentages have been added to chapter 4.

I would again like to thank Professor Tolochko for pointing out the important features of the nanocomposites. These properties were out of the scope of the current study, but are planned to be included in future work which is based off the findings.

3. ***Tensile test showed that samples incorporating CNTFs show a piezoresistive responses similar to the samples measured with standard silver electrodes (gauge factor between ~2-12), whereas the samples with metallic electrodes showed higher responses at lower force values. However, it is not clear what variations of metallic electrodes had been used. What was the main principles of metallic electrode selection?***

Response:

I would like to thank Professor Tolochko for pointing out an ambiguity in the writing of the dissertation. In the dissertation, only one type of electrode is classified and referred to as metallic, and this is the embedded copper wire used throughout the work. Effectively, the silver glue used as the reference electrode system may also be considered metallic, since the main constituent of this glue is silver. Since the silver glue was unable to perform as an embedded electrode, only the copper wire is referred to as metallic.
The selection of both of the electrode systems was based off previous literature and applications, as well as adjustment of the electrodes for this particular study. Essentially, the electrical testing of CNT/polymer nanocomposites should be done according to ASTM D257 and its adjusted variations. In the standard, a particular size and shape of a material with an insulating nature is placed between two conductive plates and tested for electrical properties using DC current. The field of nanocomposites often requires the testing of mechanical properties, and this standard is used in a widely accepted modified form where electrical contacts are "painted" using conductive adhesives around a sample periphery or at sample edges [14–16].
The copper wire, referred to as metallic electrodes in the dissertation, were selected keeping two factors in mind. The first was that they should represent metallic materials which have been used previously in literature for the purpose of measuring, imparting or forming contacts for multifunctional properties. The purpose of the dissertation was to ascertain the sensing performance of the CNTFs while providing a comparative data set to materials which are generally employed. This was chosen in order to solidify the scientific value of the findings. Works which have used metallic wiring for this purpose include, but are not limited to, [17–23]. The second factor was to use wiring of similar physical measurements to those obtained with the CNTFs in order to ascertain the effect on mechanical properties. By choosing wiring of a similar diameter, we remove variations in the experimental system which may be caused by size of the inclusions. Thus, a single diameter of 100-150 µm was chosen, which is ~5 times smaller than one data set of CNTFs, and roughly at the average diameter mark of the other CNTF data set.
In order to clarify the principles of electrode selection, additional text has been added to the dissertation in chapter 3.

I would once again like to thank Professor Tolochko for identifying this ambiguity and helping to make the text and work clearer and more understandable.

4. ***Author suggests that his concept, which is cheaper when compared to alternative techniques, has a strong potential to positively impact industrial production techniques for smart, self-diagnostic and multifunctional nanocomposites. However, this is not clear and also there are no any economical calculation.***

Response:

Professor Tolochko has again pointed out a valid concern, which has been missed in the dissertation. Currently, composite monitoring techniques for manufacturing and post-manufacturing monitoring do not overlap. The techniques for both cases include, but are not limited to visual, acoustic, infrared imagery, ultrasonic, and X-ray inspection, as well as online FTIR, NMR, DSC, and TGA are used [24,25]. Although the price range for these techniques and devices varies, they are usually on average between 1500 – 15,000

USD. This excludes any overheads that are associated with device operation. Embeddable sensor alternatives are relatively cheaper, with different systems costing between 1-1000 USD for sensors, with associated equipment for their operation costing 1000-3000 USD. For the CNTFs focused on in this study, the cost of production calculated in our laboratory is between 3-5 USD, with the multimeter options ranging from 50-800 USD, depending on sensitivity requirements.
As the comment has pointed out, the dissertation was lacking in providing some economic values associated with the implementation of this technique. Hence, additional text has been added into chapter 5 to address this. I would again like to thank Professor Tolochko for his positive contribution to this dissertation.

5. ***The work contains many references to the functional or multifunctional properties of composites, but in general, only electrical conductivity and strain sensitivity were studied.***

Response:

I would to thank Professor Tolochko for his comment regarding the definitions of the materials presented in the dissertation. I fully agree that the work focuses on the electrical conductivity and strain detection sensitivity of the nanocomposites, and this methodological and conceptual decision was taken to narrow the research findings. Electrical conductivity, piezoresistive response and altered mechanical properties are just some of the properties considered to be functional, especially for engineered materials [26]. Since these properties may be tailored to give different responses to different set of stimuli, I believe that they may be considered functional and multifunctional. Although testing for these properties is only done in two conditions (mechanically unloaded and uniaxial tensile loading), the same properties can be affected by stimuli such as more complex mechanical loading, influences of heat and humidity, as well as others. This has been shown for similar materials previously [27,28]. Furthermore, I would like to add that the polymer nanocomposites have also shown variable thermal and conductivity as reported in one of the publications as part of this thesis [6]. However, these properties are currently out of the scope of the current dissertation and are planned to be published as a separate work in the near future.

Once again, I would like to thank the Professor for his input and time which has helped polish the dissertation and made it clearer and more understandable.

Comments from Professor Dmitry Lyubchenko:

1. ***Twisting effect – what is that and how does it affect the composite properties? More explanation is needed.***

Response:

I would like to thank Professor Lyubchenko for asking a technical question which has not been clearly elaborated on in the dissertation. The process of "twisting" is a mechanical densification technique which is often employed with CNTFs. The technique relies on placing the as-produced CNTFs within a specialized holder consisting of two parts (as shown in figure 6, holders are orange in colour). The holders tightly grasp the CNTFs, and to aid this, cardboard cutouts are used at the edges of the CNTFs. Once securely seated, the two holders rotate in a counter-directional movement, one moving clockwise and the other counter-clockwise. The rotation speed and step are controlled by an electronic motor, connected to a computer, to remove variations which can be caused by manual actions. The number of twists that CNTFs can endure usually varies, and the value of 120 twists was set because fiber breakage was haphazardly seen after this value.
During this process, the CNTF edges spin at the point of being held, causing the rest of the CNTF to twist, overlap and wrap around itself. The process causes a transmittance of compressional force onto the length of the CNTF, which eventually causes a decrease in fiber diameter. This decrease in diameter caused by compression causes the voids and defects of the CNTF to be removed by essentially pressing the fiber material together. The technique has been detailed in previous works by this laboratory [29] as well as others for CNTF production and densification [30].
The technique was investigated during the beginning of the dissertation work when initial materials for study were being selected. Although the densification did seem to cause an increase in electrical conductivity, which may have been conducive to reducing contact resistance, it was found that this effect was only calculative in nature. Twisting caused a decrease in the diameter of the CNTFs, but the actual resistance readings before and

after twisting showed an increase. When these values were used for conductivity calculations using the standard formula of resistivity of solids, the smaller diameters led to higher calculated conductivity. From this, it was understood that although smaller fibers may be produced with this technique, their electrical conductivity may not actually be correctly calculated using this method. Hence, these fibers were not investigated as embedded electrodes for the nanocomposites. Only as produced wet-pulled fibers were studied. Generally, considering only the dimensions, a smaller CNTF would have led to a lower degree of defect induction in the nanocomposites [31]. However, the tighter packed surface, as seen in figure 7 of the dissertation, may have led to a decreased infusion of the CNTF and reduced interfacial interaction and adhesion.

I would again like to thank Professor Lyubchenko for posing this comment, since it indicated that the dissertation has not clearly stated that twisted CNTFs were not investigated as part of the nanocomposites. To clarify this, additional text and explanations have been added into chapter 3..

2. ***What is the physical reason of decreasing the conductivity vs. CNTF diameter in Fig.18?***

Response:

Professor Lyubchenko has posed an astute question regarding the dynamics of CNTF formation, the data for which is shown in figure 18. The wet-pulling technique was developed by the Laboratory of Nanomaterials, and the dynamics of the technique have been previously published [32]. In the technique, a thin film of SWCNTs is dry transferred onto a substrate, after which it is wettened with a volatile solvent. By doing so, a layer of liquid is formed over the film, which provides a certain surface tension. By pulling the film away from the substrate in the presence of the solvent, the film is subjected to directional forces caused by the surface tension which result in it twisting and folding, eventually causing a fiber-like structure to be formed. Immediately after twisting, the solvent begins to evaporate, causing the CNTF to densify through reduction in pore size associated with CNT bundle movement during evaporation.
In figure 18, it can be seen that the lower the diameter, the higher the conductivity of the CNTFs. Here, the conductivity of the fibers is associated with CNTF packing degree. This degree is in itself dependent upon the degree of folding and twisting of the film during the pulling process. For films of smaller width, the wet-pulling technique has been shown to be more effective in causing this densification [29,32]. In physical terms, this means that CNTFs produced from films of a lower width are able to fold and twist into fibers more efficiently, and that the solvent densification process brings about a greater degree of densification. Both of these factors cause better connections between the CNTs constituting the film, resulting in higher electrical conductivity for the CNTFs with smaller diameters.
Relevant changes in the text have been made to explain this, with changes being inserted in chapter 4, subchapter 4.2, paragraph 3.

3. ***Fig.23 tendency of changing is not clear, why?***

Response:

Professor Lyubchenko has made a valid comment concerning the tensile piezoresistive tendencies of the materials under study. In figure 23 (a) and (b), the MWCNT nanocomposites show a clearer tendency as compared to figure (c) and (d) which concern SWCNT nanocomposites. For the MWCNT nanocomposites, the embedded metallic electrodes show a consistently higher piezoresistive response as compared to measurements from standard silver glue and CNTF electrodes. Even so, samples of the same type do show slightly different behavior at different points. These variations in piezoresistive response are associated with several factors which include differences in sample dimensions, minor differences in electrode placement, stress distribution within the samples, variations in inherent electrical properties and area of final fracture. The most contributing factors in the case of this study are variations in sample dimensions, the associated differences in stress caused by this and variations in electrical properties (as per figure 22/electrical resistivity measurements after curing, between 0.2-0.5 of measured order of magnitude).
The SWCNT nanocomposites on the other hand, suffered severe mechanical property degradation with the addition of SWCNTs, resulting in a change in the base polymer from brittle to elastic/viscoelastic. For such materials, the mechanical testing is often unpredictable and varies from sample to sample, which is evident from both figure 23 as well as figure 24. Although the same factors mentioned previously contribute to the

unclear tendency, for these samples the mechanical properties of the samples is responsible for unclear trends.

4. *Fig.27. why curves are so “noisy”?*

Response:

I appreciate Professor Lyubchenko for pointing out the behavior of the various electrode systems in figure 27, which displays the cyclic piezoresistive response of the nanocomposites under study. The overall cyclic behavior displayed by the materials corresponds with the tensile piezoresistive response noted. The embedded metallic electrodes show a higher degree of response as compared to the embedded CNTFs and standard silver glue. Since an exceptionally high force value (60% of the measured average ultimate tensile strength) was used with a testing rate (10 Hz), the cyclic loading most likely caused microcracks or non-critical plastic deformation to develop within the material. Of course, like any material, the samples produced for this dissertation contained some form of defects in the form of porosity, trapped air bubbles and defects associated with molding. Such defects, when combined with agglomeration, which is noted in the SEM imagery, and agglomeration location, can cause strong peaks or valleys when monitoring a material piezoresistively under dynamic conditions. These phenomena are well noted to cause variation of piezoresistive behavior from baseline values [33,34].

5. *Tendencies for 2 and 4 contact measurements are different. Is it due to the contact resistance?*

Response:

Professor Lyubchenko has pointed out one of the key behavioral differences seen during the dissertation work, and I would like to thank him for raising this question. The Professor is indeed correct, the tendencies seen for the 2- and 4-point methods were extremely different. I would like to point out here that in the initial phases of the work, both 2- and 4-point measurements were made to determine if any differences in their measured values existed. Once it was determined that differences did exist, but only for the embedded metallic electrodes and not to the same degree for the standard silver glue, only the 4-point measurement technique was used for piezoresistive tensile and cyclic testing. The 2-point technique was only employed during manufacturing stage measurements.
In figure 19 (mislabeled previously as 29, and now corrected) and 20, the tendencies shown by measurements collected from the two techniques are visible. As the dissertation identified that contact resistance exists for the embedded metallic electrodes due to poor interaction at the microstructural level, spikes in resistance values during the manufacturing stage coupled with values which are 1-2 orders of magnitude higher than those collected from CNTFs are seen. Furthermore, even through the 4-point technique managed to remove the spikes in values (indicating that these are due to contact resistance), they are unable to completely remove magnitudinal manifestations of the contact resistance.
However, since this question was raised, it is apparent that the dissertation has not communicated this properly. Relevant changes have been introduced into chapter 5.

Again, I thank Professor Lyubchenko for raising this question and helping to make the dissertation more understandable.

6. *In Conclusion section - “During the manufacturing cycle, the CNTF electrodes showed………” Is the cycle a proper word, if yes, how many cycles of manufacturing were carried out?*

Response:

Professor Lyubchenko has pointed out a valid flaw in the writing of the dissertation with his comment and question, for which I thank him. The word cycle has been changed to “manufacturing stage” to remove ambiguity and be more precise in meaning.

I thank the Professor once more for pointing this out for correction.

7. ***Page 84, last sentence – "Comparatively, metallic embedded electrodes have the disadvantage of high noise", - is it due to the contact issue? Explanations are needed.***

Response:

The Professor has once again pointed out an area in the manuscript which could benefit from better writing quality. The sentence quoted is from the conclusions section, chapter 5. Here, I completely agree with the comment that an explanation is required, and the Professor is correct in pointing out that the embedded metallic electrodes have the disadvantage of high noise and magnitudinal values due to the presence of contact resistance.
In order to clarify this, and make the conclusions section more precise concerning the findings of the dissertation, additional text has been added to the same paragraph.

Here, I would sincerely like to thank the Professor for his time and input, and the constructive comments which have helped make the dissertation more relatable, understandable and scientifically sound and accurate.

Comments from Professor JI Puguang:

1. ***There are several images which are incorrectly numbered. This needs to be corrected.***
2. ***Images where insets are provided are extremely difficult to make out. Please consider a solution for these.***

Response:

The Professor has indeed pointed out a flaw in the thesis, and I would like to thank him for bringing this to my intention for correction. Image numbering has been corrected, as well as images being replaced with higher quality versions.

I would like to once again show my appreciation to the Professor for pointing out this mistake.

3. ***Why were masterbatches selected for study rather than powder alternatives? Is this a property and performance-based decision?***

Response:

The Professor has raised a valid question regarding the selection and usage of CNT masterbatches, for which I would like to thank them. Masterbatches were selected for study for a couple of reasons, which are as follows:

1. Masterbatches provide an added layer of safety during nanomaterial handling since they prevent the aerosolization of CNTs during the nanocomposite manufacturing process.
2. Previous works and supplier data sheets showed that with other experiments, there is no loss in property development when they are employed instead of powder alternatives [8].
3. Our own experimentation, as part of one of the works included in this thesis [6], determined that there are no major differences in functional property development when compared to powder alternatives.
4. Masterbatches are attractive to industrial applications since they provide an easy integration material form. By using masterbatches for this dissertation, I hope that the work becomes more in line with industrial interests and applications.

Hence, the decision to use masterbatches instead of powder forms of CNTs was based on both safety as well as performance factors. Once again, I would like to thank the Professor for their question and the chance to elaborate on the methodological choices made during this dissertation.

4. ***Why was DIC used instead of physical extensometers during tensile testing?***

Response:

I would like to thank the Professor for his astute comment regarding the mechanical testing procedures of the nanocomposites. Although physical extensometers are usually used as standard practice, their use was both difficult and detrimental to piezoresistive testing in this dissertation. Physically, since the electrode systems were within the gauge length of the samples, where extensometers are applied, their attachment interfered with the piezoresistive measurement contact setup. Previously, it was seen that physical extensometers, which also require electrical connections to operate, may also cause irregular readings due to short circuit creation [1]. To avoid these issues, and since a digital alternative was available, it was opted for.

Once again, I thank the Professor for his question. To add clarity to the dissertation regarding this question, additional text has been added in chapter 3.

5. ***Where do you see that possible improvements in the sensing performance can be made and how can performance be optimized or enhanced? This should be added into the relevant section of the thesis.***

Response:

The Professor has asked a very in-depth question, and provided the opportunity to showcase where and how the investigated materials have practical applications, for which I am grateful. Improvements in the sensing performance of the CNTFs can be made through a variety of changes. Some of these changes include:

1. Tailoring of densification degree: Different polymers may have different viscosities, and the optimum densification degree should be found to allow for better infiltration and interfacial interaction.
2. Precursor film thickness: By changing the thickness of films used to produce the CNTFs, the electrical properties and densification degree may be altered. These properties should be chosen and optimized to fit the needs of the stimuli being tested for. Generally, films which result in CNTFs with a lower degree of densification should result in CNTFs with higher sensitivity.
3. Additional procedures such as doping: Chemical doping of CNTFs has been shown to improve their electrical properties [29]. Appropriate chemical groups may be used to functionalize the CNTFs to make them more sensitive to particular types of nano-additives or polymer matrices.

The question posed by the Professor is quite valid to the direction of work in the dissertation and additional text has been added to chapter 5 which showcases future directions of research and possible improvement.

Again, I would like to take the opportunity to sincerely thank Professor JI Puguang for the time and effort they have dedicated to help identify issues with the dissertation and the opportunity they have provided for their correction.

Comments from Professor Alexander Kvashnin:

1. ***There are minor issues that can be addressed to the figures related to measurements (Figures 22,23,24, 27,28). As there are similar measurements for different types of materials it would be much clearer if the ordinate axis will use a consistent scale for all plots within the same measurement type to facilitate visual tracking of changes.***

Response:

I thank Professor Kvashnin for pointing out errors with the figures which have caused them to be difficult to understand. His comment has helped improve the quality of the dissertation for potential readers. In the case of figures 22-24 and 27-28, a compromise was needed since the figures display materials with difference ranges of response and testing force. However, to facilitate the visual tracking of changes, I have grouped together data from similar weight percentages and electrodes types as well as rescaled axis bars to be as close as possible. Further scaling to the same axis removes error bar size since the values of properties, as in figure 22, have orders of magnitude in difference.

Again, I would like to thank Professor Kvashnin for his valuable input in making sure that the data presented in the dissertation is clear, understandable and easy to track and compare.

Comments from Professor Alexey Yashchenok:

1. ***Chapter 3, Section 3.5.2: This chapter aims to give the main methods and materials that were used for sample fabrication and characterization. However, after reading the chapter, it seems that there is discussion of the experimental results along with the analysis of data. This somewhat make a confusion. It would be beneficial transfer discussion and analysis of data to related chapter of the thesis.***

Response:

Professor Yashchenok has made an extremely valuable comment regarding the structure and consistency of the dissertation work. The Professor is indeed correct that chapter 3, section 3.5.2 does contain experimental results and discussion. During the dissertation compilation, it was thought that perhaps adding the techniques written about in this section to chapter 3 would have been appropriate since their general findings were used for further methodological development and experimentation. However, as the comment has stated, it would be much more beneficial to include this into the next chapter dealing primarily with the results and discussion. Hence, the results and discussion of material bulk density alteration have been shifted to chapter 4 for better structure and consistency.

I would like to thank Professor Yashchenok here for provide valuable input which led to an improvement of the dissertation structure, consistency and overall quality.

2. ***Chapter 4, Page 65: It would be informative give the description how mechanical properties, such as UTC, gauge factor, etc., were estimated.***

Response:

The Professor has once again pointed out omissions in the dissertation concerning the abbreviations and terms used, and I would like to thank them for their direction in where to clarify these. To make the changes more consistent with the flow of the dissertation, the mechanical properties of interest and how they were estimated have been added into chapter 3, which goes into detail regarding the characterization techniques and equipment used. The calculative method for the Gauge Factor has also been placed in the same chapter.

Professor Yashchenok's comments have proven to be especially helpful in making the dissertation easier to understand, and once again I would like to show my appreciation for his constructive input.

3. ***Figure 28: The quality of the figure should be increased, since the insets are difficult to read.***
4. ***Chapter 4, Page 53, Figure 19: Please revise figure number.***

Response:

I appreciate Professor Yashchenok pointing out that images are difficult to understand and that a figure numbering issue exists. This comment has been made by several other reviewers as well. The quality of all figures has been improved as well as numbering issues removed.

I thank the Professor once again for pointing out omissions on my part and helping to improve the quality of the dissertation work.

5. ***Chapter 4, Page 71, Figure 27: From the figure it is not clear, why the time laps is short for CNTFs? Are they destroyed?***

Response:

Professor Yashchenok has made an astute observation and asked a very relevant question. In Figure 27, the cyclic response of the nanocomposites measured using various electrode systems is shown. The cyclic response noted from the embedded CNTFs is similar in terms of time laps to those obtained from the standard silver glue electrodes. This only cyclic response which is seen to be different is the response obtained from embedded metallic electrodes. The microstructural analysis has shown that these electrodes form a poor interface with the CNT nanocomposites as well as act as stress concentrators. Since the cyclic testing was conducted at a relatively high UTS value (60%) as well as a high cyclic testing rate (10 Hz), the plastic deformation taking place in the sample is most likely concentrated at the nanocomposite/electrode interface. These plastic deformations, often referred to in literature as microcracks, may result in a decrease in connection with the percolation network. The remaining connections are often those which are stronger than the rest, i.e. are able to sustain more deformation before breaking. These connections are often less susceptible to change in properties to stimuli, in this case being mechanical loading. Hence, the response of these samples may be seen to be delayed or not as consistent in sensitivity as the alternative materials studied. Furthermore, since the Keithley multimeter used for taking electrical measurements supplies a constant current automatically, it may need additional adjustment time to the changing number of connections. This may also lead to what is perceived as a delayed response.

The Professor is again thanked for their valuable question, which has provided an interesting aspect of the obtained results. In order to include this into the dissertation, additional text has been added to chapter 4.

6. ***From the test is it not clear that is the thickness of the carbon-based nanocomposite with embedded electrodes.***

Response:

I would like to thank Professor Yashchenok for yet again pointing out an omission in the dissertation. The thickness (3.6 - 4.1 mm, depending on nanocomposite type), and additional dimensions of the samples for mechanical, electrical and piezoresistive testing have been added into chapter 3 for clarity.

Professor Yashchenok's questions and constructive comments have indeed helped to make this work a better form of the previous version, and I thank him for the time, effort and energy he has spent to help make this happen.

Comments from Professor Yulia V. Ioni:

1. ***How was the selection of CNT film widths grounded?***

Response:

Professor Ioni has asked a valid question concerning the selection of precursor films, for which I thank her. The films were selected with different widths in order to produce fibers of different diameters. Previous work by the laboratory has compiled values for this, which were referred to [32]. These values were repeated with this work and expanded to provide CNTFs with a substantial diameter difference.

I once again thank the Professor for asking a valid question regarding the methodology of the work.

2. ***Why was the twisting selected as the densification method?***

Response:

The Professor has again asked a question regarding the methodological choices in the manuscript, and I thank her for pointing out that the justification of this choice was omitted in the dissertation. Twisting was chosen as the densification technique for a few reasons, which are as follows:

1. The technique has been previously employed by the same research group and the required experience and devices were at hand [29].
2. Other densification methods which involve chemical techniques could alter the baseline properties of the CNTFs.
3. The technique was the least time consuming to employ for initial experimentation.

Once again, I thank the Professor for pointing out that these justifications have been missed in the dissertation. Relevant text to address this has been added into chapter 3.

3. ***The work contains some errors and inaccuracies. For example, in the list of references in reference 10 there is an error in the word "filaments", in reference 44 a space is missing, some references do not contain complete output data (pages are missing). There are some problems with abbreviations in the text, for example, there is no definition of the abbreviation UTS, DIC, and, on the contrary, the abbreviation STEM does not appear in the text. Also, the author should capitalize the term "Young's modulus" (p. 15), the name of the Keithey multimeter and Intron 5969 instruments (p. 46). On the page 51 the end of the last sentence is missing and on the page 82 the author has an error in the text ("Fig. 1").***

Response:

Professor Ioni has provided extensive comments regarding omissions and inaccuracies in the writing of the dissertation, and I sincerely would like to thank her for bringing these to my attention. The references have been redone using a different software and the previous errors have been removed. The missing abbreviations have been added into the Abbreviations section of the dissertation as well as provided in their full form in the dissertation where they are first mentioned. Small definitions for these terms have also been added. The abbreviation for STEM was not added into the dissertation since the technique was not employed. Capitalization for terms as well as names of the instruments has been corrected. The incomplete sentence on page 82 has also been completed.

I very sincerely thank Professor Ioni for pointing out these omissions and mistakes in the dissertation. Her thorough examination and evaluation of the text has helped greatly to improve the quality of the work.

4. ***Figure 9 shows SEM images of three SWCNT powders of different types (briquette, pristine, RESS), but there is no SEM image of the masterbatch.***

Response:

The observation made by Professor Ioni is indeed correct and very astute. Figure 9 does show SEM images of the different powder types initially investigated. SEM imagery of the masterbatches was not deemed necessary for three reasons in particular:

1. As part of previous studies [1,2], SEM analysis of masterbatches was conducted, and their microstructure was known.
2. The SEM analysis of the masterbatches is readily available online and from the suppliers since they are commercial products. The SEM imagery conducted as per point 1 matched reference images from the suppliers and further investigation was not thought to be necessary.
3. The masterbatches employed were manufactured using the same powders seen in figure 9. Combined with points 1 and 2 and previous literature where similar materials are used, the microstructure was well known [8].

However, since SEM imagery of the other precursor materials was conducted, additional SEM imagery has been added into the dissertation in the Appendix section (appendix figure 1).

I once again thank the Professor for her efforts and comments which have made the dissertation more consistent, clear and understandable.

5. ***What is the reproducibility of the technique used for producing composites with different wt% content of single-walled carbon nanotubes?***

Response:

The Professor has asked a valid question concerning the reproducibility of the nanocomposite manufacturing technique employed in the dissertation work. As per our findings, the technique provided a variance of 10-20% on average between samples in most cases [6]. However, some processing batches did show slightly higher deviations, but remained within the same order of magnitude. Although this value may seem high at first, it is comparatively a good repeatability range when dealing with homogenously dispersed CNT nanocomposites [12].

I once again thank the Professor for asking a very practical methodological question and providing the opportunity to clarify.

6. ***The work should provide standards that define the upper limit for cyclic testing of polymers.***

Response:

I would like to thank Professor Ioni for pointing out this omission in the dissertation. Relevant text has been added to chapter 3.6 to address the issue.

The Professor is once again thanked for her diligence in reviewing the dissertation and pointing out areas of improvement.

7. ***Please explain the choice of composites with 0.25 and 0.75% by weight for subsequent multifunctional property monitoring?***

Response:

Professor Ioni has once again shown that she has diligently reviewed the dissertation, and she raises a strong question regarding the methodological choices. The same question was asked by Professor Tolochko, and was answered earlier. I believe the essence of the question is the same and in order to avoid repetition, would like to refer to the answer provided previously.
I once again would like to thank the Professor for her question, the importance of which is confirmed by a second reviewer making the same comment.

8. *The figure on page 53 is numbered incorrectly. Also, Figures 19 - 21 should be changed because they are presented in low quality: the scale along the y-axis is incorrectly selected, the captions are in small font. Figure 28 should be also changed. The 2-point electrical measurements curves for the MWCNT and SWCNT nanocomposite are highly noisy, please explain the reason. Were the experiments carried out at the same time? Please describe the external conditions?*

Response:

The comments from Professor Ioni again show her due diligence during the review of the dissertation, for which I am sincerely grateful. The comments regarding the figures, their numbering, quality and axis have been addressed in previous comments provided by additional reviewers. The experiments were conducted at the same time for 5 samples of each nanocomposite and electrode type (i.e. 5 samples of 0.25% SWCNT with embedded CNTFs were tested simultaneously). Although testing multiple electrode types may have been another option, the experimentation was limited by the capacity of the Keithley multimeter (maximum 20 contacts simultaneously). The experiments were conducted in the same laboratory with ambient conditions being controlled centrally (temperatures ranged between 22-25 ºC and humidity between 30-50%).

Once again, I thank the Professor for her excellent comments, questions and constructive contributions to this dissertation. They have indeed helped to make shape the work into a more polished form.

Comments from Professor Alexey Salimon:

Firstly, I would sincerely like to thank Professor Alexey Salimon for providing a very in-depth and rigorous review of the dissertation. His comments and questions have not only helped remove inaccuracies and address omissions, but have helped to make the work more understandable, accurate and presentable. Since the comments are extensive, they have been grouped together where possible.

1. *Polymer nanocomposite – better to explain in the beginning what does it actually mean. Nanometer scaled fillers? Nanometer scaled supramolecular structure elements?*
2. *This thesis showcases the novel application … – too "commercial" beginning, while the theses are about science and research. May I ask to rephrase? E.g. This thesis is devoted to the scientific foundations of CNTFs synthesis, integration and usage in …*
3. *Page 12. Line 3 - I guess "CNT-polymer nanocomposites".*
4. *Page 13 Line 14 – I guess "if they are sensitive to different types of CNTs"*
5. *Page 15 – Young's modulus. Y must be capital.*
6. *Page 17 Line 3. ~~characterization~~ classification*
7. *Submitted – please, give details about the journal of submission. Page 6. 4. The same - please, give details about the journal of submission.*
8. *Figure 8 – scale bar!!!*
9. *Figures 12 and 13. Captions: why a, b, c , d are given as CAPITAL? Why subscripts were changed for other characters like e instead of a1. Please, unify legend in captions in Figures 11-14.*
10. *Page 46. Keithley, not keithley.*
11. *Page 53 – Wrong Figure number – must 19. Figures 19 and 20 – the conductivity was discussed in the text above, and resistance is given in Figures.*
12. *Page 54. "the measured electrical resistance values are almost identical for the same type of nanocomposite mixture as can be seen in Figures 19 and 20." Actually, the size of Figures and scale along Y do not allow to verify this argument with naked eye. Must be improved somehow.*
13. *Page 79. "e. As seen in Figure 30, the embedded metallic electrodes display poor adhesion to both the MWCNT and SWCNT" -> Figure 31.*

Response:

I would like to thank Professor Salimon for the comments regarding omission of definitions, information, and image and writing corrections. The suggested changes have been integrated into the dissertation. The comments regarding the images were recurring between Jury Members, and all images have been improved in quality and normalized as much as possible (without losing important artifacts). I concur with the comments, which overlap with those submitted by other Jury Members, and thank the Professor for his constructive input in making the dissertation a better version.

14. ***To our great regret Professor Alexei Buchachenko died. I would ask you to revise (or include an addition) the paragraph acknowledging him taking into account this sad fact to express our mutual feelings – he was a kind person to be addressed with serious respect.***

Response:

Professor Salimon, I sincerely apologize if my dedication to Professor Buchachenko was not solemn enough. As you stated, he indeed was one of the best Men at Skoltech, and I honestly would not be here without him. I have amended the dedication, and I hope it is now somewhat satisfactory, as I believe my words cannot convey properly the positive effect Professor Buchachenko had on all of us.

15. ***Although precedence for composite monitoring using CNTFs is present for both the manufacturing and lifecycle stages of polymer matrix composites, no scientific work, to the best of the author's knowledge, has addressed their feasibility for monitoring multifunctional nanocomposites and their properties such as electrical conductivity and piezoresistive response, especially those incorporating CNTs in the polymer matrix." Please, rephrase. I see a contradiction in your argument as it is written.***
16. ***Discussion on avenues which then have not been used or studied is excessive. The same about twisting (pages 33-34). Please, exclude this, or send it to an Appendix.***
17. ***Generally ! 3.5.2 must be written more accurate and in details.***
18. ***Table 2 – The title seems to be incorrect - "series of SWCNT/epoxy nanocomposites". The density as it given corresponds to powders (and it is following from the text) or very porous foam. Are your nanocomposites porous/foamed?***
19. ***Please, specify the polymers used as a base of purchased masterbatches.***
20. ***SWCNTs by OCSiAl? In the Table 1 OCSiAl was responsible for MWCNTs. Please, clear out.***
21. ***Page 47. 1mm/min on an intron 5969 -> Instron. Can you estimate strain rate in absolute units (s-1)? This is more scientific that engineering.***
22. ***Page 51. "In comparison to metallic electrodes which have a smooth surface and surface oxide layer, this type of electrode is better suited for low contact resistance readings which allows the actual electrical conductivity and piezoresistive response of the materials they are embedded in to the measured" – seems that phrase is incomplete.***

Response:

Professor Salimon has once again pointed out inconsistencies along with providing constructive criticism as to how to improve them. For comment 15, the statement has been rewritten. For comments 16 and 17, rewriting as well as shifting to appropriate sections of the dissertation has been done, in accordance with other Jury Members who stated similar objections. In comment 18, the title of the table has been adjusted to reflect the correct meaning. For comments 19 and 20, relevant text has been added to remove the ambiguity. In comment 21, this was a writing error by myself and I see the mistake that was made. I have corrected the text to traverse head speed. In comment 22, the original sentence has been editing to its intended meaning.

I thank the Professor for his valuable comments and input, the dissertation is a now better version of what it was before.

23. ***Why do you use wt.%. If wt.% are easily re-scaled to v. % - please, discuss this and give conversion scale. In the science of composites volume fraction only has physical meaning.***

Response:

Professor Salimon has made a valid comment that in composites, volume fraction has a physical meaning while weight fraction typically does not. However, given the experimentation that was conducted, it can be seen that the bulk density states of the CNTs can be varied. This poses a problem for calculating, accurately and precisely, the volume of the CNTs used. Volume calculations with powders either rely on bulk or apparent density. These values were shown to change without any changes to the dimensions of the CNTs. Essentially, the amount of CNTs that would occupy a volume in the nanocomposites did not change (as their own volume did not), but calculations for their volume would show different values. Hence, since the mass remained constant, the mass percentage was used (which is similar to a constant particle volume). This is one of the reasons mass percentage for CNTs is often used in experimental studies [35].

I once again thank the Professor for a very correct and detail-oriented comment which has helped make the dissertation more scientifically sound.

24. ***Figure 15. At the page 38 the following compositions were described: 0.005, 0.05, 0.25 and 1.0 wt. In Figure 15 the compositions seem to be different. Please, explain.***
25. ***"SWCNT/epoxy nanocomposites fabricated by the presented route possess electrical conductivities of 0.1 – 1 S/cm at 1.0 – 2.0 wt.% " From Figure 15 I see 0.05 – 5 S/cm. Please, clear out.***

Response:

Professor Salimon has pointed out an issue with Figure 15, which I appreciate his diligence for. On this particular page, I was referring to SEM imagery being conducted for the mentioned weight percentages. Since SEM is time consuming and every weight percentage would only provide incremental information, it was decided to select specific weight percentages with large gaps between them to form an understanding of the microstructural evolution. Figure 15 displays the electrical properties measured, which was done for all the nanocomposites without skipping any weight percentages. The text on page 38 has been edited for clarification.

For comment 20, the values range from 0.33 ± 0.03 to 0.67 ± 0.05 S/cm. Since a log scale was needed to show the entire range, these values may appear to shift. The values were rounded down and up to coincide with the scale, but have now been replaced in the text.

I again thank the Professor for his time and effort which has helped remove ambiguity from the dissertation work.

26. ***Page 55. "it can be seen that the CNTF diameters provide a negligible difference for both SWCNT and MWCNT mixtures, regardless of the concentration." Also questionable.***

Response:

I thank Professor Salimon for their scientific diligence with this comment. From the experimentation conducted with CNTFs of different diameters, and the number of samples and CNTFs used, the results show that CNTFs show very similar electrical measurements of the nanocomposite matrices. There is variance in the readings, but this is within the variance limits that have been noted with the standard silver glue as well. No differences in orders of magnitude exist, even though this may be the case for the nanocomposite matrices themselves. Of course, to further solidify this study, which is a proof of concept since literature on this application does not exist, further experimentation is highly recommended in the future. Our laboratory has already begun a second study to verify these findings and identify more aspects of CNTF-CNT interaction.

I thank the Professor once again for his comment on the work, and the scientific point of view presented within it.

27. ***“The CNTFs are seen to be more sensitive to the percolation network of the nanocomposite matrix, regardless of whether SWCNT or MWCNTs have been used.” Needs to be explained more accurate and details. How do you quantify sensitivity in these experiments?***

Response:

The Professor has pointed out incorrect usage of lexicon and thus, an incorrect statement. I thank him for his intensive and extensive review of the work, which has helped to identify such areas. The statement has been changed to be more accurate in terminology and explanation.

I thank Professor Salimon once again for his efforts and time in providing a rigorous scientific review of the dissertation.

28. ***Page 57. “Variance in detected values can be seen, but it is relatively insignificant and is attributed to batch to batch processing variance”. Variance of batches? Which batches? Batches of MW(SW)CNT? Please, explain accurately.***
29. ***Page 60. Again “a batch to batch variance”! What is the batch here?***
30. ***Page 61. “CNTF electrodes show no significant difference for any of the batches.” May I ask where these “any of the batches” are presented and described in the text of thesis?***

Response:

Professor Salimon has pointed out various locations where ambiguous terminology has been used and has thus caused confusion in understanding. Firstly, I apologize for this, and have corrected the dissertation in places where the term batch has been used.

Here, the term batch refers to one amount of processed nanocomposite. Due to limitations on processing machinery and measurement equipment, only 5 samples worth of nanocomposite matrices were processed at one time. These are referred to as batches. Each batch was used to create nanocomposites with one type of embedded electrode to reduce variance between samples. For the entire study, since a large number of samples was needed, nanocomposites with the same weight % of addition were made in separate batches. This of course introduces a degree of variance in properties, which is referred to as batch to batch variance.

In order to clarify this, additional text has been added in the Materials and Methods section.

Once again, I thank you for pointing out inconsistencies and helping to make the dissertation clearer.

31. ***“They provide a more accurate value of resistance since contact resistance is not present, are more sensitive to percolation networks consisting of both SWCNTs and MWCNTs, can detect variances of CNT concentration in nanocomposites and thus can be used for CNT filtration detection.” You do not show a piece of facts proving “are more sensitive to percolation networks” !!! Please, make you arguments more accurate or show the proofs! Otherwise it is wishful arguments!***

Response:

The Professor’s scientific rigor is again thanked for pointed out discrepancies in the summary of the findings at this particular location. The text highlighted by the Professor has been adjusted and written in a way which is supported by the findings of the dissertation.

I once again would like to thank him for intensively analyzing the statements in the work, and helping to remove statements not entirely supported by the findings.

32. ***Page 66. SWCNTs do actually reduce elastic mechanical performance, not MWCNTs! Please, correct your argument “For all nanocomposite samples, it was seen that the addition of CNTs results in a loss in mechanical properties.”***

Response:

Professor Salimon has correctly pointed out an incorrect statement in the dissertation. I would like to thank him for his diligence, and have corrected to statement accordingly. Once again, I sincerely thank him for helping to make this dissertation scientifically accurate.

33. ***Page 73. "Further to note is that the samples which contained embedded metallic electrodes would fail at the site of the electrode placement, which was not the case for samples containing CNTFs or samples with standard silver contacts and that they failed at a lower number of cycles as compared to their counterparts." Not clear from this which electrodes cause sooner failure. Please, clear out. "by only 1 magnitude" -> 1 order of magnitude***

Response:

I thank Professor Salimon for their valuable comment. During mechanical testing, in both the tensile and cyclic tests, sample failure was noted at or along embedded electrodes for the metallic electrodes. When microstructural analysis was conducted, it was noticed that there was poor adhesion between the matrices and the electrodes. For the CNTF containing samples, this fracture behavior and location was not noted. Rather, a high degree of adhesion and impregnation of the CNTFs was seen. Although the cyclic testing showed the same fracture behavior for the embedded metallic electrode samples and they sustained less cycles before failure, the number of cycles sustained was not what can be called significantly lower. From the results, it can be said that the inclusion of the metallic electrodes was more conducive to failure at this location of insertion.

To clarify the statement and make it more scientifically grounded in the findings of this work, the highlighted text has been adjusted. The order of magnitude mistake has also been corrected.

Once again, I thank the Professor for their constructive comments in improving the dissertation findings.

34. ***Page 75 "can provide information regarding material health" – no proofs. None of electrode has reliably shown early signs of fatigue.***

Response:

Professor Salimon has raised a very scientifically correct point, for which I am grateful. The Professor has correctly summarized that none of the electrodes systems have picked up signs of fatigue reliably. However, here, I would like to slightly shift the focus onto the base nanocomposite. Since none of the electrode systems picked up fatigue signs, or consistently higher piezoresistive response, this is most probably behavior associated with the nanocomposite matrices themselves. For the CNTFs in this study, both linear and non-linear response to mechanical loading has been shown when plastic deformation of the fiber begins [29,36]. In this case, there is the possibility that since failure was not occurring significantly close to or within the monitoring region of the electrodes, or that the nanocomposite itself did not show such a response, that such behavior was not seen in the measurements. For the metallic electrodes, although measurement drift could be expected as they displayed contact resistance due to poor adhesion, it may have been that the contact resistance was greater than the rate of drift for stiff samples (MWCNT).

Given the correct comment by the Professor, the text has been adjusted to reflect the scientific findings of the work.

Once again, I sincerely thank Professor Salimon for pointing out such statements in the dissertation.

35. ***Please, consider to discuss more practical applications – which products and how will be tested or monitored? How to develop a protocol for CNTFs' embedding? Do you expect any local peculiarities in readings if hole, or notch, or macrocrack will occur close to a CNTF electrode?***
36. ***Do you expect some special effects if tension (quasistatic) will be compared with compression, shear, torsion, or complex loading? Please, discuss this in appropriate place.***

37. Do you expect any additional benefits if DC current measurements will be replaced with AC current measurements at a range of frequencies? Which effects may appear in AC tests?

Response:

Once again, I personally and sincerely thank Professor Salimon for providing the opportunity to highlight the strengths of the dissertation.

Further practical applications of the method outlined in the dissertation will be most attractive where hierarchical fiber reinforced nanocomposites incorporating CNTs will be used. As the CNTFs showed that they provide electrical responses which may help determine the concentration of CNTs in composites, this field would be the first place where they may be applied. Fiber reinforced composites are often subject to the filtration effect during manufacturing, and having a one-step solution for dual stage monitoring will be both feasible and attractive. The Professor, who is much better versed in these materials, may know that such composite parts are a mainstay in lightweight, high performance applications such as aerospace wings and fuselages, wind turbine structures, composite structures for construction as well as high pressure composite vessels. With the proper protocol of embedding, which would ideally be a grid like format which is premade on the fiber reinforcement, holes, notches, macrocracks and microdamage (if the grid size is small enough) should be theoretically detectable. Particular forms of damage may be detectable due to the unique range of change of resistance, but more accurate and precise equipment will be required.
In the case of other forms of mechanical loading, such as the ones mentioned by the Professor, the piezoresistive response of the base nanocomposite will dictate the response detected by the CNTFs. Generally, for complex mechanical deformation, the piezoresistive response is generally the same over a monitored area, where the resistance increases with deformation. Compression is one of the only examples where piezoresistive response for such nanocomposites is negative, i.e. the conductivity is seen to increase with compression since the percolation network is forced together. The CNTFs should, by all results seen in this dissertation, be able to pick up such responses. This has been added into the dissertation.
AC measurements should not affect the performance of the CNTFs, as they themselves have a high conductivity and will show as typical resistors (resistance value will deviate at high frequencies but at low frequencies be similar to what is seen for DC values obtained). In the case where AC measurements may be used, the base nanocomposite properties will determine changes in behavior. For highly conductive nanocomposites where the majority of connections are ohmic in nature, the behavior described above is expected. Nanocomposites which are resistive in nature will show a frequency dependence, due to the fact that the tunneling mechanism may be affected. However, this is highly dependent on many factors such as loading percentage, alignment, and nanotube type to name a few.

I thank Professor Salimon for his very extensive set of comments and questions, all of which have been extremely constructive and have made the dissertation improve in quality. I hope that the changes and restructuring are of a satisfactory quality and nature.

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