Skoltech
Skolkovo Institute of Science and Technology

Thesis Changes Log

Name of Candidate: Mariia A. Zhiliaeva

PhD Program: Materials Science and Engineering

Title of Thesis: A novel straightforward wet pulling technique to fabricate carbon nanotube fibers

Supervisor: Professor Albert G. Nasibulin

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

Dear Jury Members,

Thanks a lot for your reviews with questions as well as comments and suggestions on how to improve my thesis.

Please find below both the detailed description of modifications made in the thesis final version and answers to the questions, comments. The pages in answers are mentioned according to the final version of the thesis.

Based on the Jury member Report from Professor Alexei Buchachenko, there have been made changes according to the following suggestions:

1. *I feel a missing connection between the materials sections (3.1-3.3) and application sections (3.4, 3.5). While the former characterize a range of fibers manufactured in slightly different ways and possessing different properties, the latter stress more on the design. It would be instructive to point out what particular fibers were used (at least) for force sensor and how improvement of the fiber properties affects the sensor performance. And learn the experience on what is more important and beneficial – optimization of the fiber properties or sensor design? [The author may wish to consider revision of the thesis text in response.]*

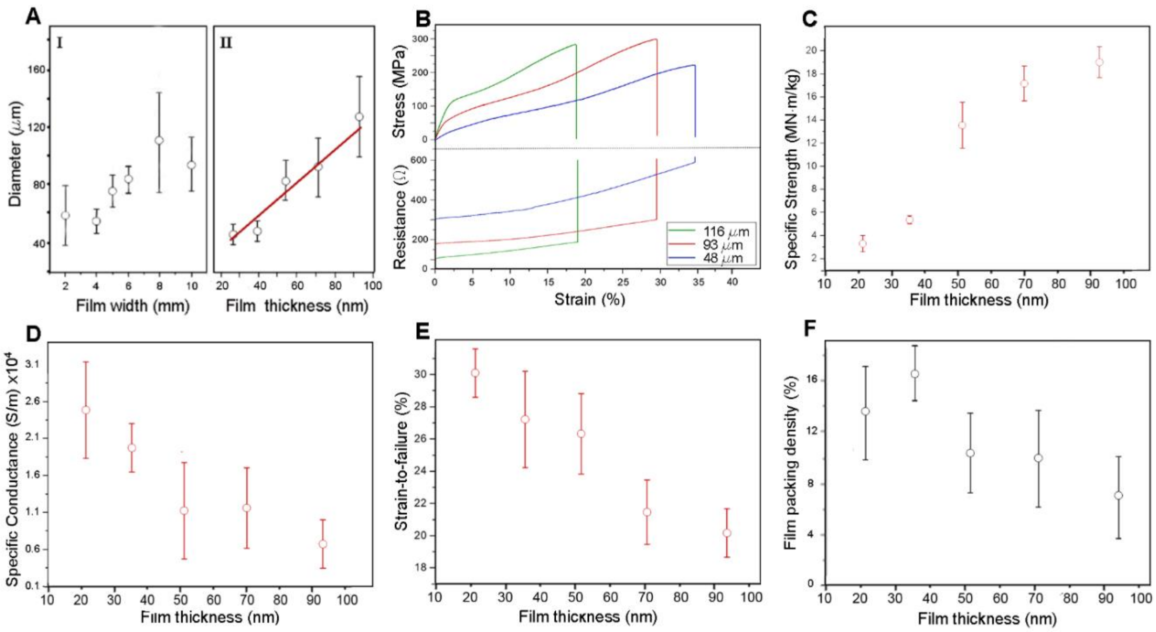
   Answer:

   Chapter 2.4 now points out the particular fiber properties for the force sensor and other applications.

   For pulse measurements, fatigue tests, and three-point bending tests we use fibers, made from a 2 mm wide strip with a 53 nm thickness. (this information was presented in the original version of the thesis (page 20, chapter 2.4)

   For all sensor experiments, we used fibers made from a 5 mm wide strip with a 93 nm thickness SWCNTs film (this information was added to the thesis, chapter 2.4).

In addition, fibers parameters recommendations are added to chapter 3.2.1. They would help to choose the right fiber characteristics for a given application.



"Figure 13. WP produced SWCNT fiber properties. (a) Dependence of the fiber diameter on (I) width and (II) thickness of initial SWCNT film. (b) The stress-strain plot and simultaneously measured resistance for different fibers. (c) The specific strength of the SWCNT fibers, made of different thickness films. (d) The specific conductance of the SWCNT fibers made of different thickness films. (e) The strain of the SWCNT fibers made of different thickness films. (f) Film packing density in fibers, made of different thickness films.

As you can see from Figure 13E the strain-to failure is greater for the thinner fibers. Thus it would be beneficial to use thinner fibers to detect/measure displacements. Thicker fibers would be better for high sensitivity, low bending applications, for example, nanophones."

"Optimization of the fiber properties may increase the sensors' sensitivity. On the other hand sensor design optimization would be more beneficial for sensing range enlargement." – added to chapter 3.4.1.

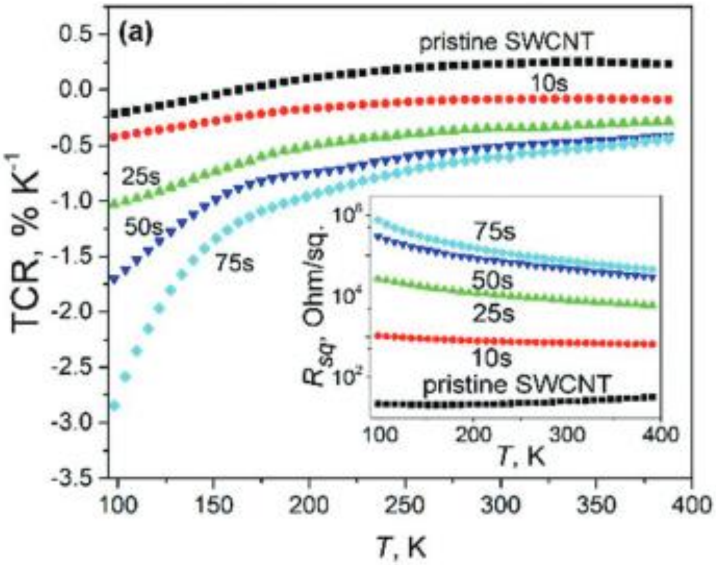
2. *Various sensing applications with fibers require quite distinct temperature regimes. Even wearable electronics should operate at temperatures higher than the room one, not to mention outdoor force or vibration monitoring. It would be interesting to learn on the stability range of fibers and how their useful properties vary with temperature. [I understand that this question is beyond the scope of the thesis, but feel that it may add an interest to in vivo discussion at the defense.]*

Answer:

According to *Kopylova, et.al.* [1] the temperature coefficient of resistance (TCR) of the pristine SWCNTs is 0.2% K^{-1} at room temperature.

For example, commercially available force sensor FlexiForce™ Standard Model A101 has a temperature sensitivity of 0.36 %/°C (according from datasheet https://www.tekscan.com/sites/default/files/resources/FLX-Datasheet-A101-RevJ.pdf)

As a result, WP SWCNT fibers (and sensors based on them) possess competitive (or even better) TCR.



The TCR response of the pristine and holey SWCNTs. (from *Kopylova, et.al.*[1])

3. *It can be more instructive to merge Tables 1 and 2 and give a short statement on the recommended way of manufacturing (perhaps depending on application targeted, see comment 1 above) [The author may wish to consider revision of the thesis text in response.]*

   Answer:

   Thanks for your comments. Table 1 represents the main fiber's figure of merit – resistance, and its changeы according to the application of different solutions/treatments. Whereas Table 2 shows various fibers parameters and covers twisted fibers parameters. Thus these two tables cannot be merged because they contain no intersected data.

   Table 2 shows that $HAuCl_4$ treatment drastically increases the specific elastic modulus and specific conductivity of the fiber. Thus it is better to utilize $HAuCl_4$ treated fibers in applications, where their conductivity is quite important, for example as passive electronic components.

4. *Subsection 3.1.3 proposes a scheme for large-scale fiber production technology. I wonder whether or not some more solid background behind (preliminary project, prototype, cost estimation, commercialization plan, etc.) does exist. [This is perhaps for in vivo discussion at the defense.]*

   Answer:

   The large-scale manufacturing scheme demonstrates only a possibility of roll-to-roll WP CNT fiber manufacturing. Its detailed design, prototype, cost estimation, commercialization plan is beyond the scope of the thesis, but an object of future research.

5. *The majority of the proposed applications lacks the comparison with alternative solutions. [Perhaps rightly, as instructive comparison would require optimization and quantitative testing. Take it as a comment for a future.]*

Answer:

It is true, that instructive comparison requires optimization and quantitative testing and would be the object of future research. Nevertheless, I added a competitive Figure 1 and Table 3 to the thesis, to understand the general performance of the fiber itself and force sensor applications to compare to others.

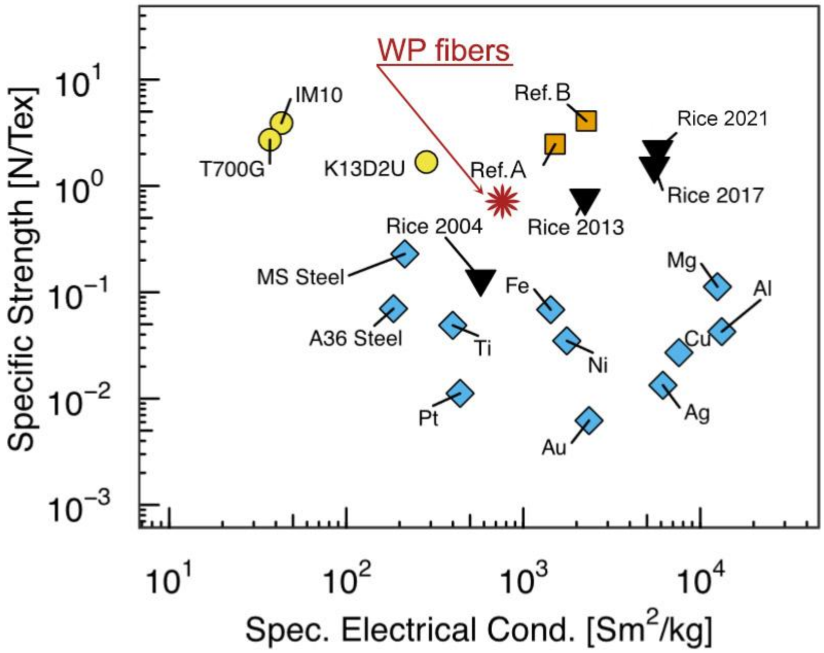


Figure 1. The best current materials performance. Based on *Taylor et.al.* [2] analysis.

Figure 1 is based on *Taylor et.al.* [2] analysis. Ref. A[3] and Ref. B[4] – are the best nowadays CNT fibers results. Rice 2004, 2013, 2017, 2021 – results obtained at Rice University over the years. Yellow rounds are carbon fibers, blue rhombs – metals.

Table 3. The summary of performance of flexible strain sensors fabricated using carbon nanomaterials in the form of membrane structure, based on review *by Yan et al.* [5].

Materials	**Methods**	**Strain range* (%)**	**Gauge factor**	**Cycle number**	**Ref**
SWCNT fiber	CVD and Wet pulling	8	14	10 000	This study
Single CNF	A microelectromechanical system	1.5	2.55	3	[6]
CNT fiber	CVD and twisting	500	-1.3	100	[7]

Silicone rubber/MWCNT	The coaxial wet spinning	100	0.68	10 000	8
Graphene/PU fiber	A layer-by-layer assembly method	50	86.9	100	9
Carbon black/natural rubber/PU fiber	A layer-by-layer assembly method	1	43.2	10 000	10
CNF fiber/PU	Embedding	2	>1700	300	11
SWCNT wire/PDMS	Embedding	12 1.1	10 000 1	5 000	12
PU/Cotton/CNT fiber	Core-spun yarn	40	-	300 000	13
SWCNT/MWCNT/PU nanofiber	Coating	50	1.24	2 000	14

* Strain range here is determined according to the fatigue test for repeatability

6. *Layout of the thesis text should be improved (broken tables, page breaks between the figures and figure captions, empty page, etc.). [This is perhaps the only obligatory revision to be made.]*

   Answer:

   Broken tables, page breaks, empty pages and other layout defects were corrected in the thesis layout.

Based on the Jury member Report from Professor Dmitry Dzhurinskiy, there have been made changes according to the following suggestions:

1. *Section 3.4: The description of FEA analysis and results should be discussed more in details. Especially what was an assumption behind modelling of SWCNT and therefore material model and specific constants used to perform the simulation.*

   Answer:

   FEA analysis was mentioned in Figure 23, page 55, section 3.4.1. It is a FEA of the elastomer sensor structure without CNT fiber. It was made to demonstrate the working principle of the sensor (how it locates, where it bends, where the fiber locates inside it; also to understand how the structure bends according to the exact load (to predict range boundaries of the applied force)). To avoid misunderstanding, the description was changed:

   "FEM analysis <u>of the elastomer structure (with no CNT fiber inside)</u> was conducted to investigate the stress-strain behavior of the sensor architecture as deflection is introduced. Simulations were carried out with Solidworks® Simulation to observe the failure deflection threshold for reliable performance. The result is presented in **Figure 23**.

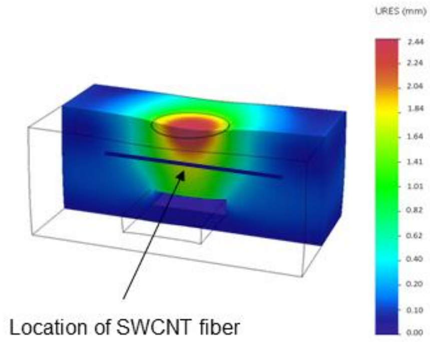


Figure 23. FEM simulation of sensor geometry with a color stress intensity gradient across its mid-section depicting the deformation due to beam-deflection controlled analysis. Location of SWCNT fiber was added to the figure after the simulation to demonstrate the fiber location inside the elastomeric structure."

2. *Since this work has potential practical application, there must be a discussion presented on the repeatability factor of the fibers produces by the wet pulling method. Especially, discussion about developed technique limitations at the industrial scale.*

Answer:

A repeatability factor of the WP CNT fibers can be evaluated from the plots in Figure 13. The following discussion was added to chapter 3.2.2:

"Error bars on Figures 13A, C, D, E, and F demonstrate the repeatability of the fibers' characteristics relative to the initial film parameter.

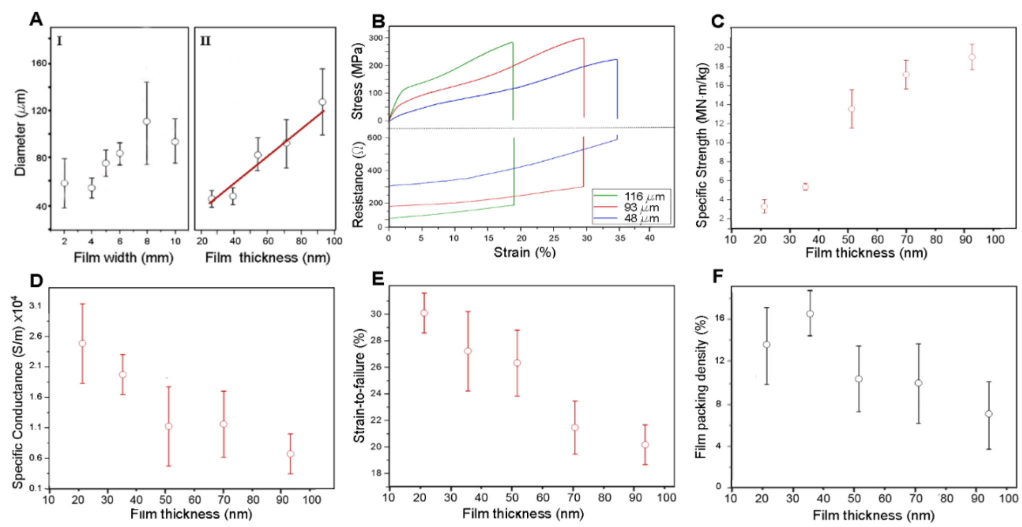


Figure 13. WP produced SWCNT fiber properties. (a) Dependence of the fiber diameter on (I) width and (II) thickness of initial SWCNT film. (b) The stress-strain plot and simultaneously measured resistance for different fibers. (c) The specific strength of the SWCNT fibers, made of different thickness films. (d) The specific conductance of the SWCNT fibers made of different

thickness films. (e) The strain of the SWCNT fibers made of different thickness films. (f) Film packing density in fibers, made of different thickness films.

It is worth noting that all presented fibers were produced manually. There are obvious factors, e.g. natural handshaking, speed, and angle variation while hand pulling, which affect repeatability drastically, due to manual operation. No doubt during industrial fiber manufacturing, where all these procedures would be automated, the repeatability factor would significantly decrease.”

Based on the Jury member Report from Professor Dmitry Gorin, there have been made changes according to the following suggestions:

1. *Page 12- 16, Introduction part should be expanded or review part should be added analysis state of the art in this research field including the figures and the demonstration more clearly the gap which the author is going to fill based on results of the research in the frame of this PhD thesis;*

Answer:

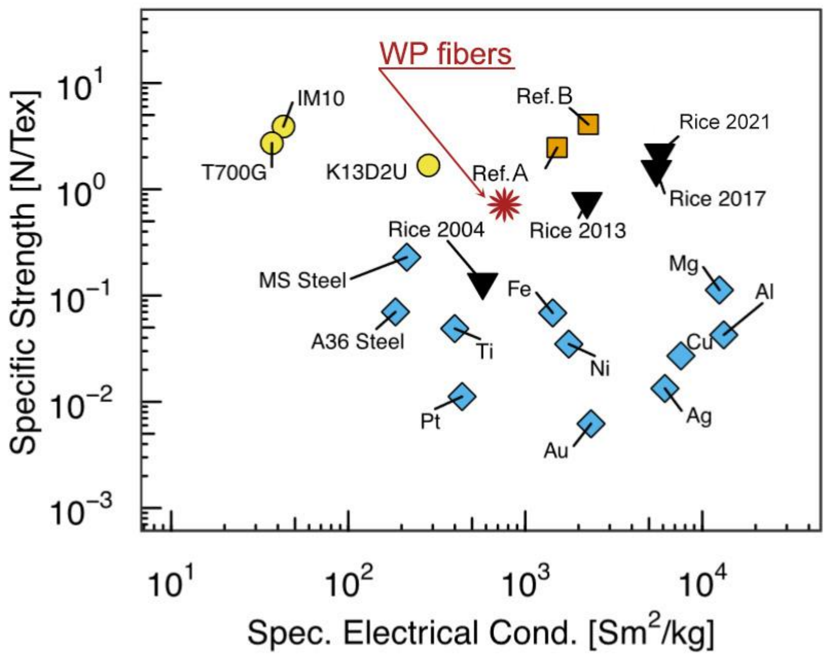


Figure 1. The best current materials performance. Based on *Taylor et.al.* [2] analysis.

Figure 1 is based on *Taylor et.al.* [2] analysis. Ref. A[3] and Ref. B[4] – are the best nowadays CNT fibers results. Rice 2004, 2013, 2017, 2021 – results obtained at Rice University over the years. Yellow rounds are carbon fibers, blue rhombs – metals.

Figure 1 shows that the WP fibers possessed compatible properties. Since it is the very first presentation of the WP fibers, there is a great potential to enhance their properties by the WP process optimization and different treatments/doping/twisting (see how Rice’s fiber’s properties have been improved over time).

Therefore, the fabrication of CNT fibers is a complex process with many requirements and limitations, which need to be simplified to open daily applications. All methods require time-consuming and complex procedures, while there is a need for a straightforward way for rapid prototyping and fabrication, which has the potential to be scaled up.

All these comments, including picture were added to the thesis.

2. *Page 23, Figure 5, Please add the time unit (second, minute, etc.) for the time axis on spectrogram and audioform and name a), and b), respectively;*

Answer:

All corrections were made.

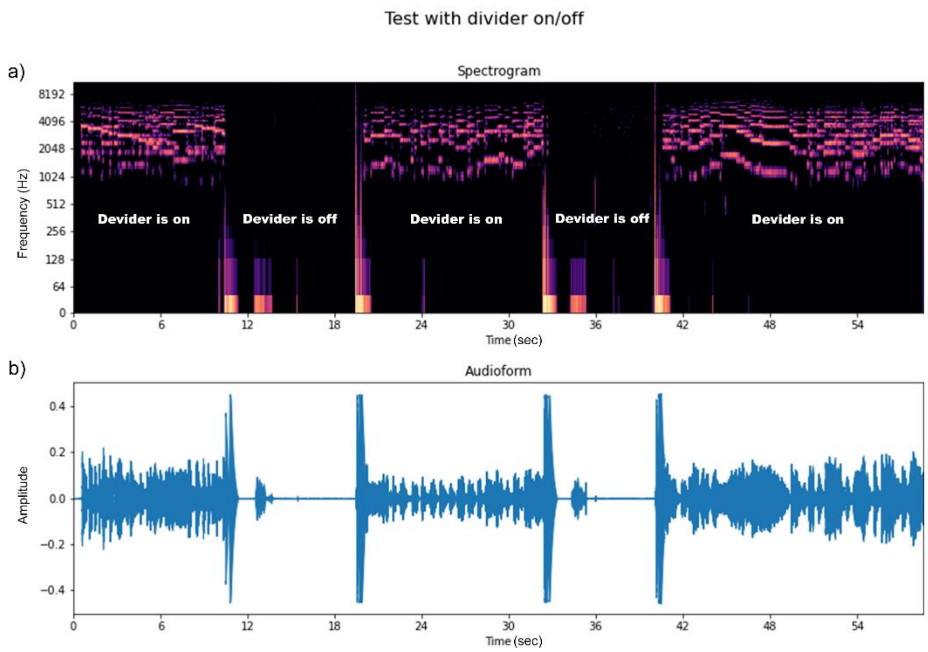


Figure 5. Acoustic test with divider turned on/off: (a) spectrogram; (b) audioform.

3. *Page 32. Figure 12, RBM on Raman spectra didn't describe. Moreover, the carbon nanotube diameter can be evaluated using the following equation d = 224/(ωRBM − 14)[M. C. Hersam, Nat. Nanotechnol. 2008, 3, 387–394, T. Jawhari, A. Roid, J. Casado, Carbon 1995, 33, 1561–1565, A. Yashchenok et al, Adv. Funct. Mater. 2010, 20, 3136–3142];*

Answer:

Figure 12 (Raman spectra) was changes to the more detailed one.

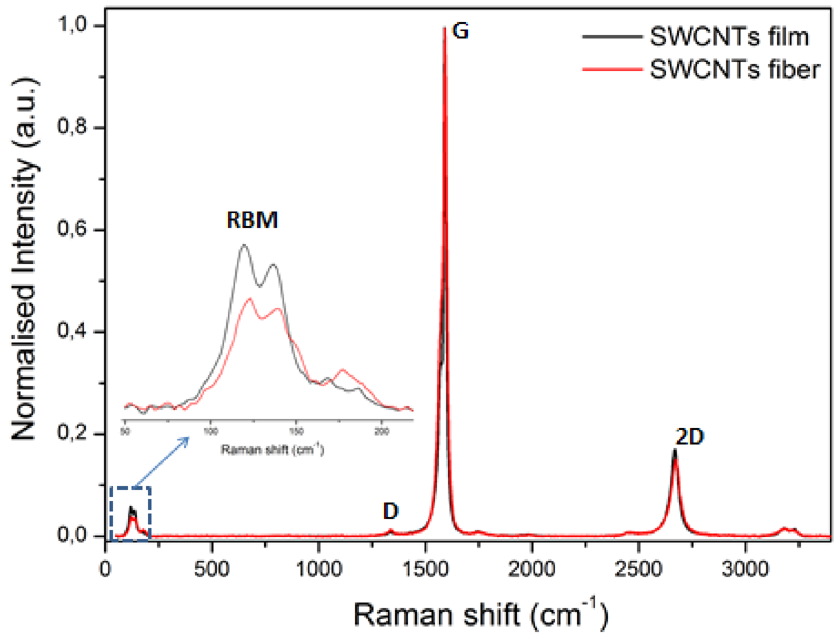


Figure 12. Raman spectra of the SWCNTs before and after the fiber fabrication.

Average diameter of SWCNTs was assessed using formula: d = 224/(ωRBM − 14). We got quite close values: 1.76 nm for SWCNT films and 1.78 nm for SWCNT fibers.

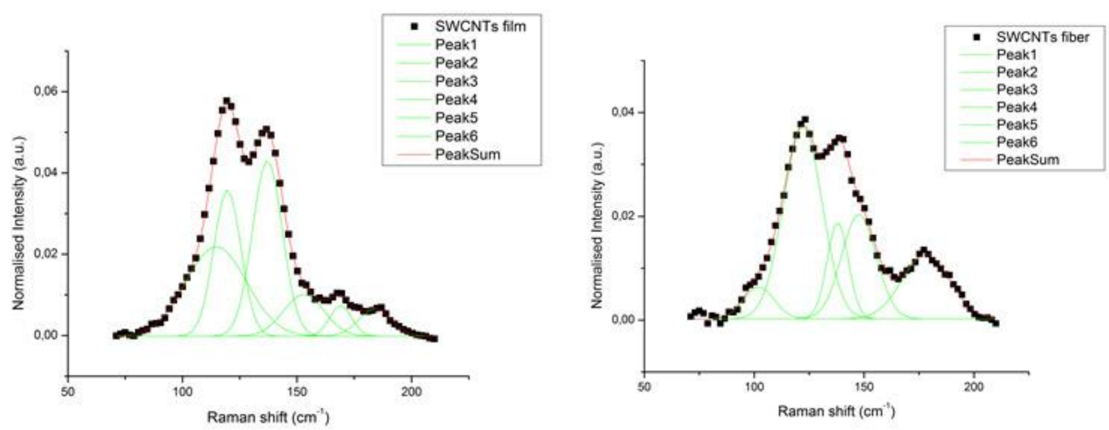


Figure: RBM analysis for SWCNT film and fiber

Film		Fiber	
Center wRBM, cm^{-1}	Diameter nm	Center wRBM, cm^{-1}	Diameter nm
114,77	2,22	102,07	2,54
119,62	2,12	122,08	2,07
137,09	1,82	138,00	1,81
152,98	1,61	147,64	1,68

169,10	1,44	178,13	1,36
184,11	1,32	197,32	1,22
Average diameter, nm	**1,76**	**Average diameter, nm**	**1,78**

Raman spectroscopy allows to determine only those SWCNTs which are in resonance with wavelength of the laser used for studies. The diameter determination using this formula is not accurate for mixed chiralities. Raman spectra was recorded using 532 nm wavelength laser, which is sensitive only for specific chiralities (see Energy-RBM frequency plot below). For more accurate diameters distribution assessment white Raman or measurements with several (continuous) wavelengths are preferred.

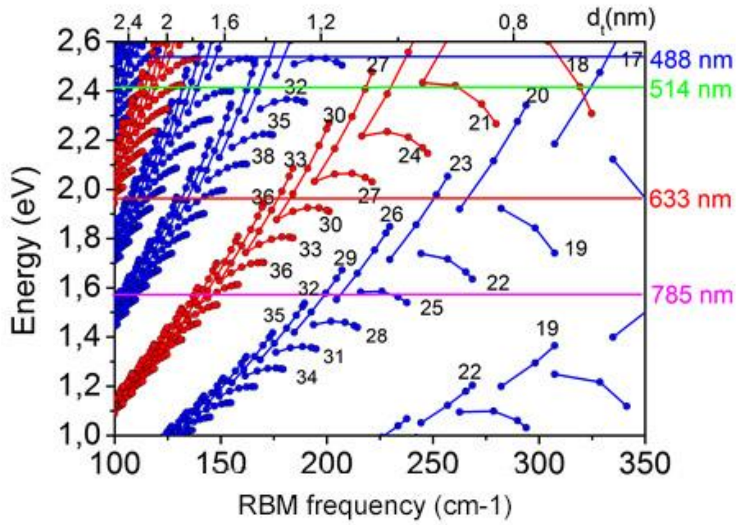


Figure: Energy-RBM frequency plot below

4. *Page 63, Figure 31, please add the time unit for the time axis for all spectrograms and audioforms;*

Answer:

All corrections were made.

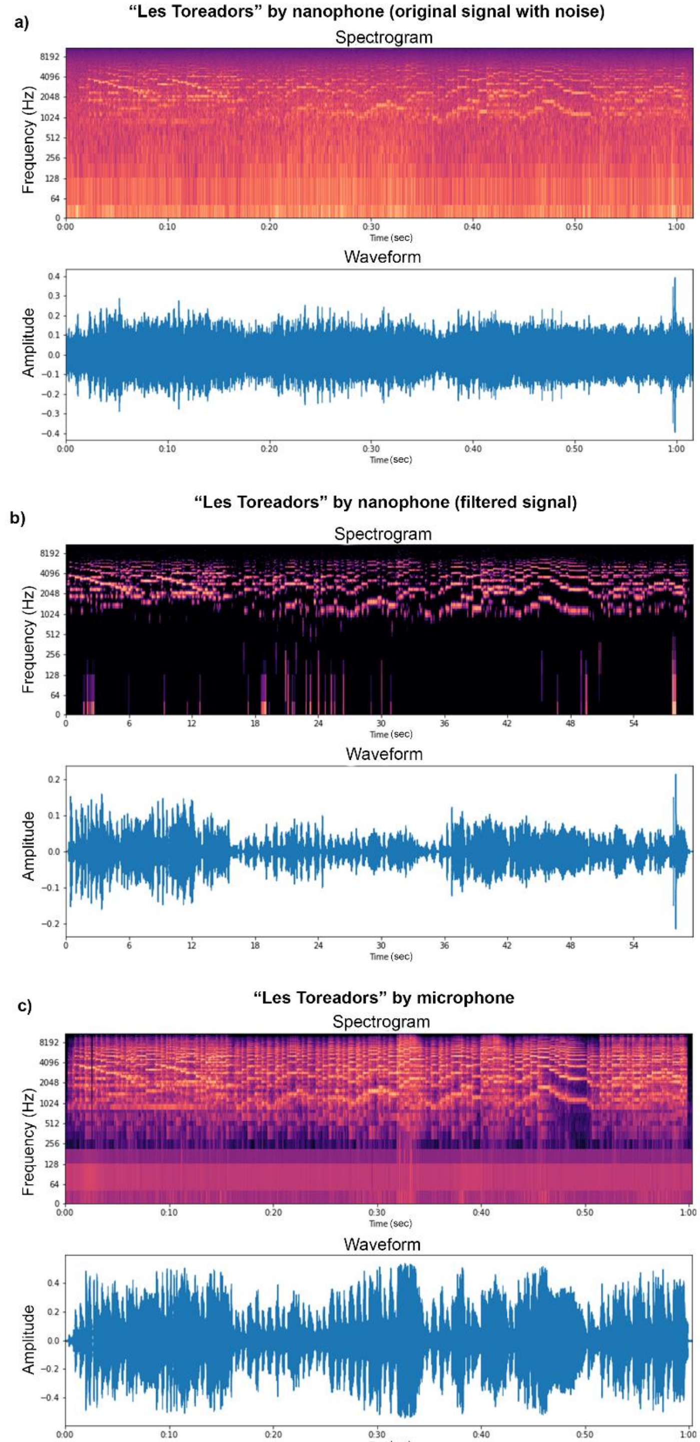


Figure 31. "Les Toreadors" recordings by nanophone and microphone. (a) Original signal of "Les Toreadors" recorded by nanophone, its spectrogram and the waveform. (b) The spectrogram and the waveform of the filtered "Les Toreadors" signal, recorded by nanophone. (c) The spectrogram and the waveform of the filtered "Les Toreadors" signal, recorded by a microphone.

5. *The author used HAuCl4 for carbon nanotube modification that can induce the gold nanoparticle growing on the carbon nanotube surface therefore I recommend to apply backscattered electron scanning electron microscopy (BSE SEM) or/and TEM for characterization of carbon nanotube surface.*

   Answer:

   Thanks for the comment! Indeed, the phenomenon mentioned by prof. Dmitry Gorin is well known for our tubes doped with gold chloride and has been widely investigated including the Laboratory of Nanomaterials, where the work was carried out. For instance, *Goldt et. al.* [15] described decoration of nanotubes' surface with metallic gold nanoparticles with a size of approximately 5–10 nm (Figure a and b) after $HAuCl_4$ treatment at temperatures up to 300 °C. For samples thermally treated at 400°C, in addition to outer surface decoration with Au^0 nanoparticles, we can notice the filling of the inner SWCNT space (Fig. 3c and d).

   Measurement of the interplanar spacing of the encapsulated material from the TEM images resulted in a value of 0.235 nm. It was admitted to be metallic gold, which has a inter-planar spacing of 0.2355 nm. Gold nano-particles decorate the outer surface of SWCNTs, forming via spontaneous reduction of $[AuCl4]^-$ anions. When the heat-treatment temperature is high enough to oxidize the nanotube caps, e.g. 400°C, the doping solution penetrates into the SWCNT inner space, resulting in improved doping and, also, the formation of the metallic gold phase. This is manifested in higher doping efficiency for the SWCNT film treated at 400°C.

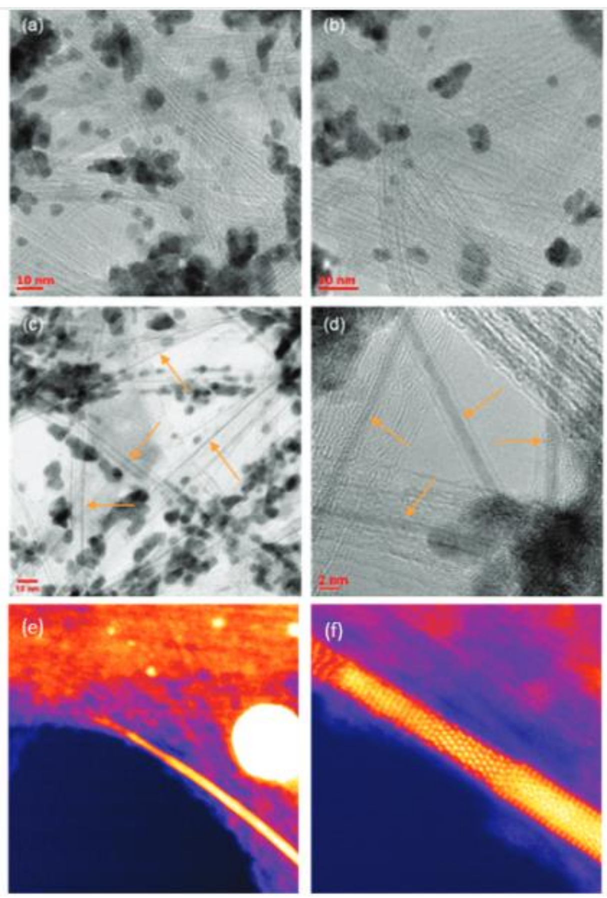


Figure: TEM images of SWCNT films doped with 15 mM $HAuCl_4$ ethanol solution: without preliminary thermal treatment (a); pre-treatment at 300°C (b) and 400°C (c and d). Arrows indicate SWCNTs filled with the metallic Au phase. STEM images of 15 mM $HAuCl_4$-doped opened SWCNTs with encapsulated gold nanowires (e and f). (from *Goldt et. al.* [15])

Based on the Jury member Report from Professor Tanja Kallio, there have been made changes according to the following suggestions:

1. *Chapter 3.1 describes wet pulling technique applied to randomly oriented SWCNT and CNT dispersion. Details on the fibers made of aligned CNTs are missing. Would the fibers' characteristics stay the same if they would be made of aligned CNTs films? Would it be possible to make the same devices based on them?*

   Answer:

   Of course, it is possible to apply WP technique to any kind of CNT film. But different CNT films' properties lead to different CNT fibers' properties. If the film would be stronger/more conductive the resulted fiber would be stronger/more conductive. As for aligned CNT fibers, all fibers' characteristics would correlate with the film characteristics.

   It is not possible to make sensing devices out of aligned CNT fibers. The ability to "sense" comes from the random orientation of the CNTs inside the film. During stretching, CNTs rearrange, and the resulting resistance changes. In the case of aligned CNTs this effect would be negligibly small (1) and irreversible (2), because (1) there is always some level of not alignment and (2) such CNT film stretching results in cracks, but not rearrangements.

2. *Figure 8, Chapter 3.1.3 describes a potential large-scale production line. Chapter 2.2 describes using solvent and dopants during the wet pulling procedure (WP) to enhance fibers properties. Can they be applied during this roll-to-roll manufacturing process?*

   Answer:

   Yes, they can be applied. All of them can be applied during stage III in Figure 8. This comment was added to chapter 3.1.3 of the thesis.

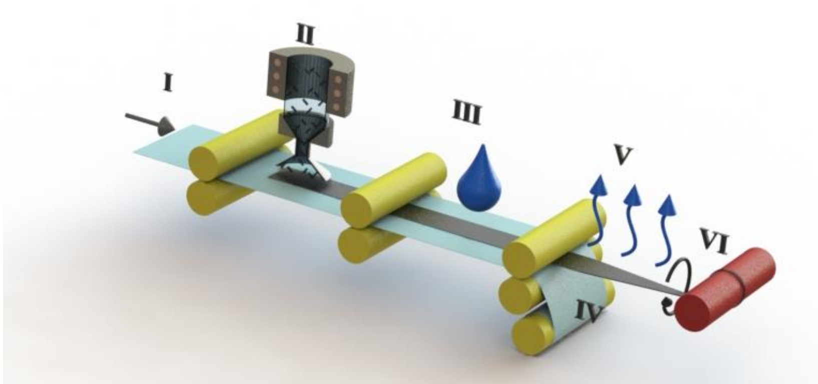


Figure 8**.** General scheme for the WP CNT fiber production. I) Filter supply; II) CNTs deposition on a filter; III) Solvent impregnation; IV) Filter separation; V) CNT film folding and solvent evaporation; VI) Coil the CNT fiber on the bobbin.

3. *Chapter 3.3.1 describes PDMS encapsulation. Which other materials can be used for encapsulating WP CNT fibers?*

   Answer:

   We utilized PDMS since it is stretchable, transparent, and bio-compatible. Depending on the target application, elastomers, ceramics, textiles, and even composite structures can be used. This comment was added to chapter 3.4.1.

4. *Chapter 3.4 describes impressive applications, including a tunable force range sensor. What are the actual boundaries for the applied force measurements?*

   Answer:

   Changes in the resistance are induced by individual CNTs displacement related to each other. As a result, the lower boundary is limited by the resistance measuring hardware (and reduced external noise (wires, contacts, etc.)). The upper boundary is limited only by a sensor (stiffener) architecture and material.

5. *Chapter 3.4.4 describes a nanophone device based on the WP fibers. Figure 30 shows the PDMS encapsulated fiber, which is in direct contact with the speaker. Can the nanophone record the sound waves remotely in environments other than air?*

   Answer:

   Yes, it can. A nanophone is able to record sound waves in different environments. Such a hydrophone can record the sound for much greater distances. Below is the link to the records of the random knocks, made by hydrophone (underwater nanophone) from a distance of 20 cm.

   https://youtu.be/j8xRlk-QV8Q

Based on the Jury member Report from Professor Krisztian Kordas, there have been made changes according to the following suggestions:

1. *Although the text cites the contemporary literature very well, and refers to tens of papers published on carbon based piezoresistive sensors, somehow a good comparative table compiling various carbonaceous materials and device structures/assemblies with their gauge factor and applicable stress/strain window is missing. This is only a minor flaw that may be fixed by showing and discussing some related data of peers (in the light of the results presented in the thesis) during the public examination.*

   Answer:

   Table 3. The summary of performance of flexible strain sensors fabricated using carbon nanomaterials in the form of membrane structure, based on *Yan et.al.* [5] review.

Materials	**Methods**	**Strain range* (%)**	**Gauge factor**	**Cycle number**	**Ref**
SWCNT fiber	CVD and Wet pulling	8	14	10 000	This study

Single CNF	A microelectromechanical system	1.5	2.55	3	6
CNT fiber	CVD and twisting	500	-1.3	100	7
Silicone rubber/MWCNT	The coaxial wet spinning	100	0.68	10 000	8
Graphene/PU fiber	A layer-by-layer assembly method	50	86.9	100	9
Carbon black/natural rubber/PU fiber	A layer-by-layer assembly method	1	43.2	10 000	10
CNF fiber/PU	Embedding	2	>1700	300	11
SWCNT wire/PDMS	Embedding	12 1.1	10 000 1	5 000	12
PU/Cotton/CNT fiber	Core-spun yarn	40	-	300 000	13
SWCNT/MWCNT/PU nanofiber	Coating	50	1.24	2 000	14

* Strain range here is determined according to the fatigue test for repeatability

As shown in the Table 3, WP CNT fibers possess compatible characteristics with other carbon nanomaterials-based sensors. They have a potential for a greater number of cycles (we haven't made fatigue test for more than 10 000 cycles, most probably these properties remain the same even after). Given strain range and gauge factor fit most of the applications, while simplicity of the WP technique provides it an advantage for rapid prototyping.

These table and comment were added to the thesis.

Based on the Jury member Report from Professor Oleg Tolochko, there have been made changes according to the following suggestions:

1. *Chapter 3.4 presents numerous applications. I wonder what are the advantages and disadvantages of the presented force sensor compare to other existing sensors.*

   Answer:

   As it was written above, Table 3 now added to the thesis and provide a full description of the WP strain sensor properties compare to others.

Table 3. The summary of performance of flexible strain sensors fabricated using carbon nanomaterials in the form of membrane structure, based on *Yan et.al.* [5] review.

Materials	**Methods**	**Strain range* (%)**	**Gauge factor**	**Cycle number**	**Ref**
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SWCNT/MWCNT/PU nanofiber	Coating	50	1.24	2 000	14

* Strain range here is determined according to the fatigue test for repeatability

2. *Chapter 3.4.1 describes the force sensor with adjustable force range. There is a lack of details of its actual force measuring limits.*

Answer:

Changes in the resistance are induced by individual CNTs displacement related to each other. As a result, the lower boundary is limited by the resistance measuring hardware (and reduced external noise (wires, contacts, etc.)). The upper boundary is limited only by a sensor (stiffener) architecture and material.

3. *There is no any characterization of CNT used for the experiments. Is it possible to use any single wall CNTs for the sensors or there are some special requirements?*

Answer:

Yes, it is. Crucial phenomena in the WP process are film folding by surface tension and densification during solvent evaporation. In general, a CNT integrity in fibers is determined by van der Waals forces, an innate strength, and surface tension. As a result, we can utilize different kinds of CNT films, but the resulting fiber properties would also vary. In general, more conductive/stronger is an initial CNT film, more conductive/stronger is resulted CNT fiber. Moreover, only fibers made of randomly oriented CNTs can be utilized for sensing applications. Aligned CNTs inside the fiber prevent it.

4. *The text layout may be improved. There are breaks between figures and figure captions, and empty pages.*

Answer:

All corrections in the thesis layout were done.

Best regards and thank you once again,

Maria.

Bibliography:

(1) Kopylova, D. S.; Fedorov, F. S.; Alekseeva, A. A.; Gilshteyn, E. P.; Tsapenko, A. P.; Bubis, A. V; Grebenko, A. K.; Popov, Z. I.; Sorokin, P. B.; Gladush, Y. G.; et al. Holey Single-Walled Carbon Nanotubes for Ultra-Fast Broadband Bolometers †. *Nanoscale* **2018**, *10*, 18665.

(2) Taylor, L. W.; Dewey, O. S.; Headrick, R. J.; Komatsu, N.; Peraca, N. M.; Wehmeyer, G.; Kono, J.; Pasquali, M. Improved Properties, Increased Production, and the Path to Broad Adoption of Carbon Nanotube Fibers. *Carbon* **2021**, *171*, 689–694.

(3) Wang, J. N.; Luo, X. G.; Wu, T.; Chen, Y. High-Strength Carbon Nanotube Fibre-like Ribbon with High Ductility and High Electrical Conductivity. *Nat. Commun.* **2014**, *5*, 1–8.

(4) Lee, J.; Lee, D.-M.; Jung, Y.; Park, J.; Lee, H. S.; Kim, Y.-K.; Park, C. R.; Jeong, H. S.; Seung, &; Kim, M. Direct Spinning and Densification Method for High-Performance Carbon Nanotube Fibers.

(5) Yan, T.; Wang, Z.; Pan, Z.-J. Flexible Strain Sensors Fabricated Using Carbon-Based Nanomaterials: A Review. *Curr. Opin. Solid State Mater. Sci.* **2018**.

(6) Cai, J.; Chawla, S.; Naraghi, M. Piezoresistive Effect of Individual Electrospun Carbon Nanofibers for Strain Sensing. *Carbon* **2014**, *77*, 738–746.

(7) Li, Y.; Shang, Y.; He, X.; Peng, Q.; Du, S.; Shi, E.; Wu, S.; Li, Z.; Li, P.; Cao, A. Overtwisted, Resolvable Carbon Nanotube Yarn Entanglement as Strain Sensors and Rotational Actuators. *ACS Nano* **2021**, *18*, 15.

(8) Tang, Z.; Jia, S.; Wang, F.; Bian, C.; Chen, Y.; Wang, Y.; Li, B. Highly Stretchable Core−Sheath Fibers via Wet-Spinning for Wearable Strain Sensors. *ACS Appl. Mater. Interfaces* **2018**.

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