Bringing Colours to the Black Box —

A Novel Approach to Explaining Materials for Evolution-in-Materio

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***Abstract*—The work presented in this paper is done within the NASCENCE (NAnoCSale Engineering for Novel Computation using Evolution) project which investigates computational properties of nanomaterials and unconventional computing paradigms which can be applied to such materials in order to achieve material computing. The paper presents a novel approach to modelling computations in one of the nanomaterials considered in the project - Single-Walled Carbon Nanotubes (SWCNTs)-polymer nanocomposites. Our belief is that the presented approach is more suitable for the experiments within NASCENCE. It takes inspiration from the principles of dynamical hierarchies and can be related to some known cellular computing architectures. Our motivation is given as well as some initial simulation results based on a simplified model built according to the proposed approach. The results show that the proposed approach captures well the conductivity dependence on the concentration of carbon nanotubes and varying electric potential in the material.**

Keywords–Unconventional computing; Evolution-in-Materio; carbon nanotubes; SWCNT-polymer nanocomposites.

I. INTRODUCTION

Computing devices based on silicon (Si) technology are not likely to meet the needs for the extent of computations needed by humankind in the future. Novel technologies need to be found which will provide for the growing scales of computations but also for the demands such as energy efficiency and similar. In that it is not only sufficient to find a promising replacement for Si, i.e., some novel material which can perform computations; it is also necessary to discover new computing paradigms better suited to novel technologies.

The NASCENCE project [1] investigates both. Materials considered so far are coated gold nanoparticles and nanocomposites made of SWCNTs and polymer molecules but also, recently, SWCNTs / Liquid Crystal (LC) dispersions. Methods of material manipulation for achieving computation are explained within Evolution-in-Materio (EIM) [2][3] whereby Evolutionary Algorithms (EA) are employed to search for the solution of a computational task in materio.

Exploitation of computational properties of a material requires a good understanding of its properties and behaviours when exposed to certain type of excitation. A good understanding of the material properties is best tested in a model of the material. If a model produces responses similar to the experiments in the lab, then model is sufficiently good to make good predictions of the material behaviour in simulation thereby saving experimental time and resources.

However, to make a model good enough for the CNT-based materials used in NASCENCE is not an easy task. This is due to the setup of the experiments but also the bulk of material itself: there is no tidy alignment of CNTs, rather there is a blob of nanocomposite where Van der Vaals' forces keep SWCNTs in bundles which, sustained by polymer molecules, stretch in all directions. Further, there is no 'neat' electric field as is the case, for example, in the CNT-gate stretching from source to drain electrode in thin film transistors [4][5]. In NASCENCE experiments, electric field is the result of the voltages on a number of electrodes which are immersed into a blob, thick film of SWCNT-polymer nanocomposite. As existing models and simulation methods fail to serve the purpose, a novel approach is needed.

This paper presents one of the approaches considered within the project. In NASCENCE, a material under experiments is treated as a black box - some signals are brought to it and some signals are read as a response. If the aim is to model what takes place in the material, we need to unbox it and look inside searching for the physical processes which lead to material computing. Different levels of details can be accounted for when addressing the physics at the bottom of the computations produced by the material. The approach presented in this paper is based on two paradigms borrowed from complex systems: dynamical hierarchies [6][7] and cellular computing [8][9].

The paper is organised as follows. Section II gives a brief overview of the computing materials and EIM. Section III presents the approach to modeling SWCNT-based nanocomposites. Section IV shows simulation results based on a simple model and, finally, Section V provides a discussion of a proposed approach and some directions for future work.

II. MATERIALS THAT COMPUTE

The notion is not novel. Ferrous sulphate which discriminates between the frequencies [10], frequency discriminator in FPGA tissue [11], pattern recognition in a bucket of water [12], robot controllers in liquid crystal [13] - are just a few examples, some of which have been known for as long as half a century. Beside finding suitable materials, another challenge is which method of material manipulation to use in order to achieve useful computation. One possible way is to use a bottom-up approach and let an EA search the space of possible solutions. EIM [2] uses computations performed by the material when stimulated by some signals from outside material which change

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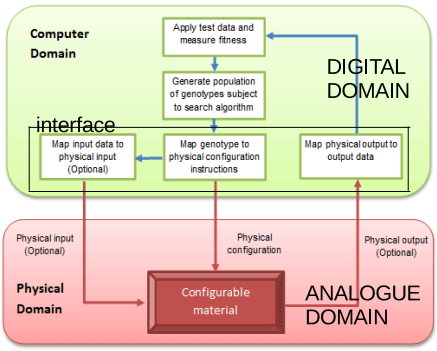

Figure 1. Overview of the Evolution-in-Materio, taken and adapted from [14]. See Section II for explanation.

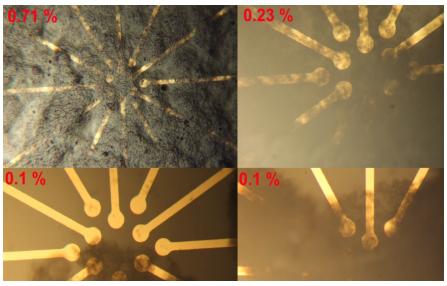

Figure 2. Optical micrographs of SWCNT-polymer nanocomposites dispersed over gold electrodes with $25\mu m$ electrode gap. Different concentrations exhibit various coverage of gold electrodes.

according to an EA. These signals configure the material so that it performs a certain computation. Therefore they are called *configuration signals*. EA is run on a computer where configuration (signals) of the material is represented as a genotype which undergoes evolutionary changes until material response corresponds to a desired computation. Figure 1 shows an overview of this process.

Configuration and input signals as well as fitness value are processed by the computer, i.e., they are digital while actual computations which happen in the material are analogue due to the physics exploited for computation. Figure 1 shows this cross-domain between digital and analogue worlds. Instructions which configure the material are generated by the computer (digital) and are subject to an EA which runs on a computer. Material response (analogue) is read and converted by interface so that it can be tested for fitness by the computer.

EIM is used in NASCENCE [1]. In a typical experiment, material is treated as a black box. We assume nothing about what is inside the sample of material when searching for solutions for some computational task. A more detailed description of the process used in the experiments can be found, for example, in [15][16].

In NASCENCE, it has already been shown that materials can solve computational problems [14][16]. However, having a problem solved is not enough. We would like to learn more about the possibilities that lie in the material. If the travelling salesman problem is successfully solved for 9 and 10 cities [14], can we expect the same blob of material to be equally successful at solving the problem for larger numbers of cities? Can we say more about problem scalability in some other way than purely by running exhaustive runs of experiments?

To get more knowledge about the computing properties of the material, a way to go is to construct a model of the material. A good model means a good understanding of the underlying laws of physics which govern behaviour of the system under investigation. A model also shows how well we understand the material - if a model is good enough, when used in simulations it will produce behaviours similar to those observed in the lab when we run experiments. The more we know about the material, the better can we make use of its properties for computations while saving experimental resources.

III. DICING MATERIAL INTO CELLS

Many models of CNTs exist [17]–[21]. However, usually they are suited for a certain purpose, for example to model conductivity of CNT gates in thin film transistors [18]. Our goal is to model conductivity of CNTs used in NASCENCE experiments. The conductivity of CNTs is due to the percolation paths they form and can be observed when CNTs are placed in an electric field. It changes either with the change of electric field or with the change of percolation paths if nanotubes are movable. In case of the samples currently under investigations, SWCNTs are not movable, they are held at certain positions by polymer molecules. So, the only way that conductivity of the sample can change is by changing the electric field to which the sample is exposed.

Therefore, it can be said that the challenge faced when modelling blobs of SWCNT-polymer nanocomposites used in NASCENCE experiments is multifaceted. The main two questions which need to be answered are:

- how to represent physics of SWCNT-polymer nanocomposites?
- how to represent electric field which changes according to the change of voltages on the electrode array?

Figure 2 shows micrographs of some of the samples used in NASCENCE experiments. Typically, it is a $20\mu L$ blob of SWCNT-polymer nanocomposite. Polymer is Poly(Methyl MethAcrylate) (PMMA) or Poly(Butyl MethAcrylate) (PBMA). The nanocomposite is dissolved in some solution to be dispersed over array of electrodes as a droplet and subsequently dried at high temperature leaving a thick film of nanocomposite spread over electrodes upon dissolvent's evaporation. Electrodes are made of gold and can be placed in different arrangements - circular, square matrix - so that the distance between individual electrodes varies. Typically, it is several 10s of μm and so is the radius of individual electrodes.

Another feature which can be noticed from micrographs in Figure 2, is that the coverage of electrodes by nanotubes is not uniform. Some electrodes are more in contact with nanotubes than the others and some may even remain with no contact at all. Also, this is very much dependent on the SWCNT concentration. A somewhat simplistic sketch of the material sample at hand can look as given in Figure 3, which simplifies the content of the sample together with the electrodes.

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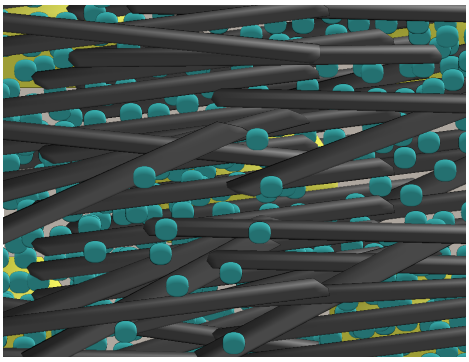

Figure 3. A sketch of the system used in experiments: SWCNT bundles are represented as grey sticks lying in all directions, polymer molecules are in a shape of balls and electrodes are golden yellow patches seen behind material.

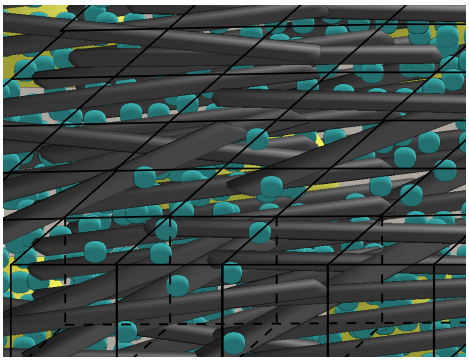

Figure 4. Example of the system viewed as a collection of cells.

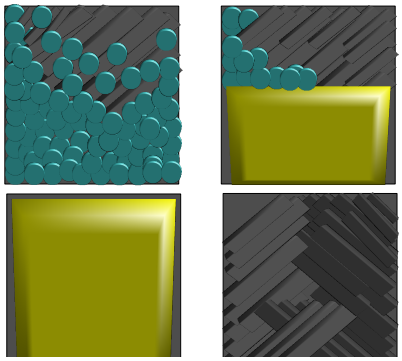

Figure 5. Four examples of what the cell sides may look like.

A. *Abstracting away the details*

Behaviour of complex systems is the result of the dynamics of their constituent parts [22]. In [23], Herbert Simon named hierarchies as "one of the central structural schemes" that the architect of complexity uses. And, indeed, hierarchies, dynamical hierarchies [6][7] in particular, describe systems which are made of a number of components undergoing some dynamics and, in doing that, mutually interacting. Such behaviour leads to the emergence of novel units at higher hierarchical levels [24]. Hierarchical systems within which higher levels emerge as a result of the dynamics of lower level units have been studied from various approaches - philosophical, computational, information theoretical - that more as they carry the principles on which life itself emerges.

Interesting to notice is that dynamics at different levels happens at different rates [25]. The lower the level, the higher the rate of the dynamics [24] which further leads to averaging or "some selective loss of detail" as Pattee names it [26] or some filtering of the information from a lower to a higher level [27]. All the details of the dynamics at lower levels need not be fully known, rather some averaged, filtered information which describes it in sufficient detail.

B. *Cellular approach*

Another trait of complex systems is that their constituent parts operate in parallel. The behaviour or functionality of the system as a whole is the result of the operation of its parts but, as a rule, it cannot be simply represented as a sum of its parts, i.e., nonlinearity is inherent to the system. The related notion is *emergence* [28] which captures the property that "the whole is greater than the sum of its parts" and that nonlinearity is present.

Models of such systems are usually based on the representation of the parts of the system as cells. It is needed to find appropriate cell arrangement and some governing mechanism for the cell dynamics. This mechanism can be given in different forms - rules which describe how a cell transitions from one state to another - Cellular Automata (CA) [8] or some physics of the cell - Cellular Neural Networks (CNN) [9].

C. *Our sample - what is in the cell?*

Now, let us take a second look at the sketch of a system shown in Figure 3. Let us try to imagine some 3-dimensional cells, in the simplest case let them be a one layer of cubes, as in Figure 4. Each cell contains a portion of the system which may contain SWCNTs and/or polymer molecules and/or golden electrodes. The cell dynamics within the system will be governed by the physics of its content. The cell interaction with its neighbouring cells will be governed by the sides where the cells touch. Figure 5 shows some examples of what the sides of the cell, in this case a cube, may look like. The top left figure shows an example with mainly polymer molecules and CNT bundles only in a right top corner; the top right an electrode with CNT bundles and a few polymer molecules; the bottom left an electrode; the bottom right only CNT bundles. If only polymer molecules are present, there will be no current flow between the cells as they are electrically isolators. If there are SWCNTs there, the current flow will be determined by the electrical properties of the SWCNTs - the percentage of metallic and semiconducting nanotubes, characteristic resistance per unit length, etc. If there is an electrode there, then the voltage it provides is known. Further, the sides of the cells may contain each of the elements to a certain percentage.

The behaviour of the cell is determined by the physics of the material the cell contains. When describing the physics that governs the cell dynamics, no detailed mechanisms of SWCNT-polymer nanocomposites behaviour are needed. Some level of abstraction may suffice so that interesting and useful behaviour is still captured although not all the segment and junction resistances and the currents in individual segments of percolation paths formed by SWCNTs are considered.

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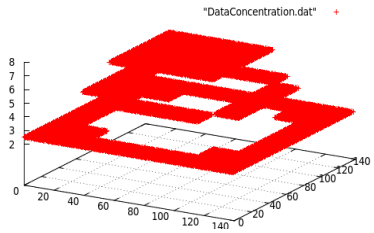

Figure 6. Plot of the concentration of SWCNTs in our model, see Section IV for explanation.

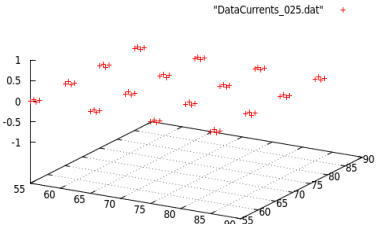

Figure 8. Plot of the simulated current for the SWCNT concentration 0.25% - only the cells which contain electrodes can conduct current but since no percolation paths are formed with other cells, there are no current flows.

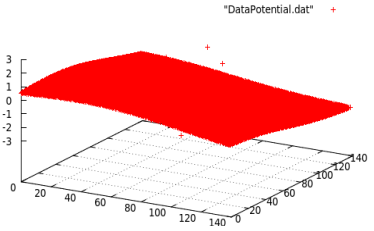

Figure 7. Plot of the electric potential resulting from the voltages on electrodes, stand-alone crosses correspond to the electrode voltages, see Section IV for further explanation.

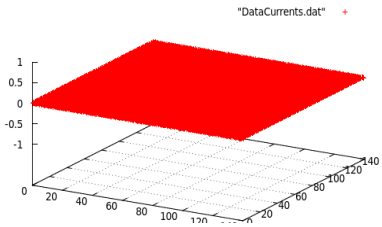

Figure 9. Plot of the simulated current for the SWCNT concentration 5% - all cells in the system conduct current.

IV. Model and Simulation

Let us consider a very simple model which captures the properties described in Section III. A sample of material is given as a blob of SWCNT-polymer nanocomposites dropped over 16 electrodes placed in 4×4 array. Further, let us assume that such system has been diced into a number of cells, as in Figure 4. The cells are uniform and in a shape of a cube. Approximately $1.96 \cdot 10^4$ cells of $100 \times 100 \times 100 \mu m^3$ dimensions would account for a blob of $20 \mu L$ of nanocomposites typically used in our experiments. Let them be placed in a 140×140 grid of cells and position the array of electrodes in the centre of the grid.

Lab samples are characterised by the concentration of SWCNT. To make the model more realistic, let us assume that the concentration is not uniform for all the cells: the cells closer to the centre of the 140×140 grid have slightly higher concentration than the cells closer to the edges. Therefore, in our model, for a given concentration, we multiply this value with some weight which depends on the position of the cell within the grid, as shown in Figure 6, for a simple case where the concentration of 0.25 is multiplied by coefficients 1.5, 1 and 0.5 dependent on the cell position within the three 'rings' around the central cells which contain electrodes, 1.5 corresponding to the closest to the centre.

Electric field is the result of the voltages on electrodes. During EA runs, voltages on electrodes change thereby changing electric field as well. The electric field at certain location determines electric potential of the location. Without going into exact equations, let us assume that the value of the electric field at some position within the 140×140 grid is given as supersposition of the contributions from individual electrodes. Further, for each contribution, the value of the electric field is dependent on the distance of the cell from that particular electrode. Figure 7 shows a plot of the electric potential caused by the electric field which is determined in the described way for voltages on electrodes being some random numbers in the range $[0, 3]V$. For each location (i, j) in the grid, the potential is calculated as $v_{ij} = 0.01 \cdot \sum_{k=1}^{16} v_k \cdot \sqrt{(i - k_i)^2 + (j - k_j)^2}$, for an array of 16 electrodes in a 4×4 matrix.

The change of the states of the cells is governed by the physics of the material found in the cell. In our model, the cell state is described as the collection of the states of the cell sides $(v_i, i_i), i = 1, 2, ..., 6$, the state of the cell side given as a pair of values (v_i, i_i) representing electric potential and current at its location within the grid. The choice to describe the state of the cell based on the states of its sides comes from the fact that conductivity of the sample is calculated based on the interactions between the neighbouring cells which can allow for the current conduction if sufficient concentration of CNTs is found at the cell boundaries so that conducting percolation paths can be formed.

Let us consider a simple case where the physics is determined purely by the concentration of the SWCNTs in the cell and some probability that the side of the cell will conduct. In particular, a random number from a uniform distribution is multiplied by the concentration of SWCNTs in the cell and if such product is greater than the midrange of the distribution interval, the current may flow and is set to 1, otherwise it is set to 0. Figures 8 and 9 show the conductance of the material for two corner cases when there is no current as no percolation paths are formed due to a low concentration of SWCNTs and when concentration of SWCNTs is so high that current flows across the whole of material.

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V. Discussion/Conclusion and future work

In this paper, initial work on modelling conductivity of SWCNT-polymer nanocomposites has been presented. The novelty of the approach lies in it being based on two paradigms: dynamical hierarchies and a cellular computation model. Principles of hierarchies are used to find a suitable approach to abstracting away details of the physics at a very low level. A cellular computation model accounts for the interactions between units of material - cells.

Initial sketch of the model shows that when it is used in simulations, it yields the results which are intuitively expected: higher concentrations of SWCNTs in nanocomposites lead to more percolation paths formed and consequently more current flows; different voltages on electrodes produce different electric fields in the material; the material behaviour is based on some underlying physics which can be abstracted to a certain level, etc.

However, this initial sketch is still far away from the model of the behaviour of the materials used in NASCENCE experiments - it needs to be extended! We identify main directions for future extensions:

- representation of the physics which drives the behaviour of the cell
- type of the cell which is best suitable

Could some of the existing models for CNT thin film transistors be used like in [19]? Or some of the models currently investigated within the project (not published yet)? Or some even higher level description? Simple cubes as presented at this place are far from the real blob of material sample. Some other shapes, e.g., dodecahedron are being considered. Also, the non-uniformity of the cells is likely to be suitable for successful models regarding size and content of the cell.

We are looking forward to addressing these challenges.

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