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Evaluation report on the PhD thesis of Ms. Ashmita Bose, MSc, entitled "Evolutionary algorithms as a tool for designing chemical computers" and submitted for the award of the degree of doctor of philosophy in the field of chemical sciences to the Institute of Physical Chemistry, Polish Academy of Sciences, Warsaw, Poland

The PhD dissertation of Ms. Ashmita Bose, MSc, focuses on developing a framework for elaboration of chemical computers as a breakthrough perspective for information processing, which challenges conventional computing technology founded on the silicon chip and it opens a new perspective for an efficient and fast spreading of information. As a model chemical environment, the Candidate selected the Belousov-Zhabotinsky (BZ) oscillatory chemical reaction. As a theoretical model of the BZ reaction, she selected a simple Oregonator model which consists of five second-order elementary steps, the corresponding kinetic equations, an autocatalytic step, and a delayed negative feedback loop. In that way, the Oregonator model fulfills with an excess all theoretical demands posed by Ilya Prigogine on description of an effective oscillatory chemical model (e.g., represented by Brusselator). Oregonator is a simplified version of a more complex FKN model of the BZ reaction, which embraces as many as 18 elementary steps. Needless to add that in chemical literature, a number of other BZ reaction models also exist, just to remind the Györgyi, Deutsch and Kőrös model (1987; 17 elementary steps), or the Försterling, Murányi and Noszticzius model (1990; 16 elementary steps), both originating from an excellent Hungarian school of physical chemistry and nonlinear chemical dynamics.

In my opinion, selection of the BZ reaction for the purpose of this study in order to develop chemical processor is ingenious, and for more than one reason. Owing to its periodicity and to a relatively long-term perpetuation of chemical elementary steps involved in the BZ reaction, it can be considered as a chemical switch model (with a theoretical potential at least) to process and transfer information. Dynamics of this oscillatory reaction can conveniently be steered in a neat external manner by using the ruthenium catalyst and illumination of the reaction vessel with blue light of different intensities, in that way manipulating oscillation frequency (and hence, manipulating performance of this chemical processor). With low light intensity, practically no effect on the oscillation period is observed and with an increased light intensity, the oscillation period increases. Surpassing the threshold value of the blue light intensity, oscillations are eventually extinguished, as demonstrated in the paper by Showalter et al. (*J. Phys. Chem.*, 1997).

Upon formulation of the concept of the BZ-based chemical processor model, the following perspectives have opened for Ms. Ashmita Bose, MSc: (i) She could either propose a detailed outline of the chemical computer architecture (employing the BZ reaction model and engaging the Oregonator reaction as a construction tool), or (b) she could alternatively perform practical experiments, e.g., on functioning of the blue light controlled BZ reaction under the different working setups. As a result, the Candidate took advantage of both options, theoretical and practical.

The PhD thesis under the discussion is composed of seven chapters (plus several addenda) structured in a traditional manner. Chapter 1 'Introduction' offers a preliminary glance at the Author's research interests to become the subject matter of her thesis. In Chapter 2 'The basic concepts of chemical media used for information processing, elements of information theory and evolutionary optimization', the Author defines the framework for her research exploration. In Chapter 3 'Computing with interacting chemical oscillators – network designing and teaching strategy', she provides assumptions of her own concept on how to build a network of chemical processors able to transmit information based on the Oregonator model. Chapters 4 and 5 ('Networks of chemical oscillators as classifiers of geometrically inspired problems' and 'Networks of chemical oscillators as classifiers of medically oriented problems') provide the results of two practical applications of the chemical oscillator-based classifier to the selected practical problems. In Chapter 6 'Towards experimental realization of a chemical computer using Belousov-Zhabotinsky reaction', Ms. Ashmita Bose, MSc, presents the results of her own practical experiment, nicely imbedded as a link in the chain of earlier attempts going in the same direction. Finally, Chapter 7 'Conclusions and discussion' summarizes and evaluates the results obtained in the area of research performed by the Author. The evaluated PhD dissertation is abundantly illustrated with several dozen of well selected figures and 123 references to the scientific literature. Last not least, Ms. Ashmita Bose, MSc, is a co-author of four research papers already published and falling within the scope of her post-graduate research interests and of one paper in preparation. In that way, she more than sufficiently meets the requirements posed on the publication output of a PhD Candidate under the Polish law and tradition.

In the theoretical part of her PhD task, the Author provides an overview of earlier experiments which have engaged the BZ reaction carried out under the specific working conditions (e.g., in very narrow tubes, in very thin layers of liquid reaction medium etc.), and which have purposely been devised in such a way as to demonstrate certain features of these systems which might suggest possession of specific "memory" and a possibility to be used for transfer of information. These features promise a chance to elaborate the BZ reaction-based chemical switches. In the next step, she makes herself acquainted with the information theory and information processing, to eventually decide for the method of neural networks for information transfer. To this effect, she first starts from modeling a single BZ reaction-based chemical oscillator and then she devises a network composed of a number of synchronized oscillators, based on the Oregonator model. To this effect, she has to face a fairly complicated issue of numerically solving the set of kinetic Oregonator equations and then to optimize kinetic parameters adequate for a given practical task. Finally, she succeeds in training a network of chemical oscillators to solve two geometrically oriented problems, one of them being establishment of a colour of a randomly selected point on the Japanese flag, and the other one being differentiation between two intertwined spirals. In both cases, the tasks are positively solved with use of the developed oscillator networks.

In the practical part of her PhD task, the Candidate continues efforts earlier commenced by her Co-Supervisor Prof. Jerzy Górecki (and also by some other research teams worldwide) in optimization of an externally controlled and effective coupling between the separate chemical processor units. For this purpose, she imbeds (following a strictly defined experimental protocol) the BZ reagents and ferroin in two porous DOWEX 50W-4X resin beads considerably different in size and then she studies an impact of the bead-size-depending distance between these two units on synchronization of oscillations running inside them. Final statement which concludes the experimental part of the Candidate's PhD thesis is that the diameter of the two DOWEX 50W-4X beads exerts no measurable effect upon synchronization, and it is the distance between the two units alone which matters (in this sense that the shorter is the discussed distance, the more rapidly synchronization of the oscillations by these two units is approached).

The PhD thesis by Ms. Ashmita Bose, MSc, is a dissertation with a noteworthy component of a far-reaching vision. In its very centre, the oscillatory type reaction is positioned, otherwise stirring a large spectrum of quite diverse emotions, which – as we all well know – can spread from an enthusiasm and awe to disbelief, or even to negation. This mistrust toward oscillatory reactions, especially those running in homogenous liquid media, is well reflected in general reservation of the international chemical community, if not in abstinence from this particular subject matter. Looking wider, this tendency is reflected in a stubborn abstinence from the discussion of dissipative structures, complex systems, and irreversibility of chemical processes in most undergraduate and graduate chemistry curricula and handbooks of the higher education enterprises worldwide. The research task predefined by the Author (guided by her two Co-Supervisors) has been completed to the full and it adds considerable importance to oscillatory reactions (often perceived as a trick or a fun fact) by laying her personal brick to the edifice of a future technology of spreading information without an aid of the silicon chips.

Finally, may I use my position as a reviewer and address the Candidate with certain tempting questions which seem unanswered in her PhD thesis and which may gain her attention in the course of the defense discussion. In view of a considerable effort which has been invested not only by the Candidate, but also by her predecessors in practical experiments targeting optimum conditions for developing chemical processors based on the BZ reaction run in a droplet (or in a porous bead rather), what are real perspectives of technical success of these efforts? In your response please, highlight the following two issues: (i) Firstly, none of chemical reactions, neither oscillatory or not, is a *perpetuum mobile*, so that it expires for one reason or another after certain period of time. On the other hand, an idea of dispensable "chemical chips" is rather difficult to imagine. (ii) Secondly, the ion-exchange resins like DOWEX (either anionic, or cationic) can affect pH of the reaction and in that way perturb reaction mechanism. Moreover, chemistry of their inner surface does not remain unchanged in the course of a prolonged action, as its functionalities gradually decay. Why the microporous glass beads with a stiff structure and an inert inner and outer surface have not been considered as an alternative material which is chemically stable, easily available, and which is employed on a routine basis as stationary phase in quite popular Size Exclusion Chromatography (SEC) technique?

At the end, I would like to express my profound conviction that the PhD thesis entitled "Evolutionary algorithms as a tool for designing chemical computers" and submitted by Ms. Ashmita Bose, MSc, more than sufficiently fulfills all formal and essential requirements posed on this type of academic output (as formulated in: Art. 187 ustawy z dnia 20 lipca 2018 r. Prawo o szkolnictwie wyższym i nauce, Dz.U. z 2018 r., poz. 1668 ze zm.) and therefore in my view, it should be accepted for the further procedural steps in its present form.

Moreover, I propose to award the discussed PhD thesis with distinction for the Author's courage and imagination to explore this area of science which is challenging much above an average, and also for the results obtained in this field. In my opinion, exploration of the oscillatory reactions is among the most challenging areas of science in general, due to the fact that research of chemical phenomena falling within the scope of the non-linear thermodynamics needs to be founded on unequivocal kinetic experimental data which are still hardly available, due to the contemporary state-of-the-art in chemical analysis. One part of PhD thesis by Ms Ashmita Bose, MSc, is dedicated to this particular aspect of carrying out kinetic investigations in the field of the non-linear chemical reactions, and namely to practical controlling in real time the rate of the BZ reaction under the employed experimental conditions.

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