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Review of Habilitation Thesis

"Macromolecules and nanostructures in a low temperature plasma" by Dr. Jaroslav Kousal

Dear Prof. Dolezal, Dear Prof. Nemecek,

you asked me by a letter from 22.02.2023 for a review in respect to habilitation procedure of Dr. Jaroslav Kousal.

Thank you for the invitation to assess the habilitation thesis. With pleasure, I report on the activities of the candidate and the originality of his thesis – and I hope that it will be helpful for the habilitation procedure. Please, find enclosed the related report.

As opponent I formulated a few questions / remarks in my report which may be used for the scientific discussion in the habilitation procedure. They are not to be seen as criticism.

Sincerely yours

Review of Habilitation Thesis "Macromolecules and nanostructures in a low temperature plasma" by Dr. Jaroslav Kousal

I know Dr. Jaroslav Kousal since many years by interesting discussions at international conferences on complex (dusty) plasmas and plasma technology as well as by his excellent papers published in related journals. During the intense discussions and joint meetings in Prague and Kiel and at workshops I recognized Dr. Kousal by his solid knowledge and exciting ideas in research of plasma-wall interaction, in particular, for plasma polymerization and nanocomposite deposition as well as in plasma diagnostics as a distinguished scientist.

Mr. Kousal is working as academic researcher since 1998, currently focused on the investigations of plasma polymerization processes for various applications, partially in space technology. After several years of PostDoc he became an assistant professor at Charles University. The special research activities of Dr. Kousal are over years in the field of diagnostics and applications of low-temperature plasmas for polymerization processes, applications of nanomaterials and magnetron sputtering processes. The colleagues of the international community in plasma physics and technology recognize, especially, his interesting work on experiments and modeling of nano-particle formation in gas aggregation sources (GAS) and the diagnostics of nano-dusty plasmas. The latter is a crucial approach for the plasma and sheath analysis in reactive and particle-containing plasmas. The response of typical plasma properties to particle formation can be used for the description of the dynamic behavior, charging processes and deposition of nanocomposites. These research aspects, combined with chemical and morphological particle and surface analysis as well as the interesting considerations of collisional reactive plasmas, have strongly supported the activities of plasma studies at Charles University initiated by Prof. H. Biederman and others.

In addition, I also would emphasize the broad experience of Dr. J. Kousal in material analysis for nanoparticles and nanocomposites by irradiation sources which is of great importance for innovative applications in plasma technology. He established and supported several collaborations on this subject with different groups in Germany (e.g. DESY Hamburg). Mr. Kousal is author or co-author, respectively, of more than 70 peer-reviewed publications in international journals and communications in related conferences. Most of the papers are published in important journals of plasma physics and material science.

In his habilitation thesis, Dr. Jaroslav Kousal selected 12 of his publications to highlight the interplay between plasma-surface interaction, formation of nanoparticles / nanocomposites and plasma polymers. Doubtless, the synthesis of these different topics in the framework of his cumulative habilitation thesis, which consists of a comprehensive introduction (or summary) and the original papers, is a challenging task successfully worked out. The selection of the related contributions as well as the guiding thread through the thesis turns out well. Since the included papers have already been undergone a peer-review process before publication in the related journals, I can only underline the originality, scientific quality and importance of them in the framework of the habilitation. I just want to highlight the pioneering work of diagnostics, characterization and optimization of gas aggregation sources (GAS) for the generation of (metallic) clusters and their subsequent deposition into nanocomposite films. Experiments and simulation on the formation of nanoparticle and their distribution in the plasma source are quite useful for understanding and optimization of the involved processes.

Of special interest are the correlation between the surface temperature of substrates or nanoparticles, which is mainly determined by the energy influx from the process plasma, and their properties (morphology, etch or deposition rate etc.). How can the experimental findings of the author (p.3) be explained by the so-called Thornton structure zone diagram recently modified by A. Anders? Why is there an increase in the etching rate of thin BSA films for 3 mm nozzle-sample distance (Fig.2.2)? Dr. J. Kousal studied in great detail the magnetron sputtering processes of polymers (Fig.2.3). What are the advantages of magnetically confined electrons and what are the problems for their diagnostics (e.g. by Langmuir probes) in such an environment? Concerning the effect of nitrogen admixture in sputtering leads to heavier molecular species during sputtering as mentioned at p.7. What are the underlying mechanisms for this observation? The discussion related to the formation of NH_x radicals (p.8) could be extended in the light of collisional processes and existing literature for such reactive plasmas – see for example, the papers of M. Sode et.al.

In addition to the interesting experimental investigations the candidate also made effort in simulation of nanoparticle formation, distribution and transport in the GAS (Fig.2.8) which is quite commendable. For the description of electrostatic deflection of charged nanoparticles in the GAS chamber empirical models have been developed (p.12). What about the different charge states (negative, positive, neutral) of the generated metallic clusters and nanoparticles? For the discussion of the transport and behavior of the nanoparticles Dr. Kousal uses methods and tools of dusty plasma research (p.14). In particular, the effect of the different acting forces (depending on the particle size) should be discussed in detail. The generation of clusters / nanoparticles in the GAS due to sputtering processes is mainly by the coalescence of colliding species (p.16). Which role play surface processes at the target (spikes, flakes) due to the change of the target state during its use? On the other hand, in addition to problems with the magnetrons magnetic field also nanoparticle formation disturbs the recording of Langmuir probe characteristics. How to handle these problems?

The discussion of plasma polymers (p.19) is quite interesting as well and considers many results and experience of the author in this research area. Again, the role of the energy

influx is very important for these processes and has been discussed by many authors (Yasuda, Hegemann, Kersten ...) as a kind of macroscopic similarity parameter. How are the elementary processes (adsorption, sticking, desorption, crosslinking, reaction etc.) are affected by the resulting surface temperature and what is the influence of momentum transfer by the incoming species for the polymer film properties? The planned improvement of the plasma-assisted vapor thermal deposition (PAVTD) by the candidate will be certainly appreciated for a better understanding and optimization of plasma-based polymerization. The perspectives for future applications (in particular, in space technology) may be outlined and described.

Just a short comment on the form of the thesis: It is clear and well written and clearly structured related to the different included original papers. Still, the equations should be numbered and the figures should be placed near the text where they are firstly mentioned and explained.

Summarizing, the selected papers together with the guiding Introduction / Summary are an essential contribution to the broad research field at the border between plasma physics, material science and diagnostics. The habilitation thesis emphasizes that Dr. Jaroslav Kousal is highly qualified for this subject and I am convinced that he will essentially contribute to the increase of knowledge and technology in this innovative topic in the future.

I can state that the thesis (summary and included papers behind) is definitely of high originality. I support the application of Mr. Kousal and I recommend the acceptance of his habilitation by the Charles University in Prague.

Kiel, 10.05.2023

Prof. Dr. Holger Kersten