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Report on the PhD thesis entitled

"*SYNTHESIS OF IRON(II) DOPED COPPER FERRITES AS NOVEL HETEROGENEOUS PHOTO-FENTON CATALYSTS*"

submitted by

Engr. Asfandyar Khan

Asfandyar Khan worked in material science during his PhD studies in the Doctoral School of Chemical Engineering and Material Sciences, Institute of General and Inorganic Chemistry, University of Pannonia. **He studied the** effect of different metal oxides ($Cu^{II}O$, $Fe^{II}O$, $Fe^{III}_2O_3$) and iron(II) doped copper ferrite ($Cu^{II}_{(x)}Fe^{II}_{(1-x)}Fe^{III}_2O_4$, x = 0, 0.2, 0.4, 0.6, 0.8, 1.0) and their photocatalytic applicability for the degradation of model contaminants/dyes. Due to potential (and already realized) applicability in nanotechnology, research activity on this topic is very high. However, it is clear to everyone who works in this field that there are still many things to develop and discover around these photo-Fenton-type systems. Thus, the choice of topic is undoubtedly actual.

The Thesis is relatively well-structured and correctly presented, which consists of five main chapters. Abstract, Table of Contents, Lists of abbreviations, Tables, and Figures are introduced at the beginning of the dissertation. The dissertation is written on 90 pages and appendixes. A brief overview of the importance of the targeted material is provided in Chapter 1 – Introduction (28 pages). A sub-chapter deals with the dissertation's research objectives/goals, which is more than necessary. However, I appreciate the development/corrections made on the original/primary version of the manuscript, I still think that the first paragraph of this section is dealing with way too general aspects of research (e.g. the description about methylene blue would fit in the Introduction of the Thesis).

The work provides a relatively comprehensive summary of the topic using 220 references, proving his deep knowledge about the field. Section 3 is dedicated to the description of the used materials and methods (10 pages). The experimental techniques and methods are also up-to-date and suitable for the fulfilment of the aims. The as-presented chapter suggests that the Candidate has learned these techniques and can evaluate the data obtained using these methods. Although, I have not found any hints as, which measurements were performed directly by the Candidate or which he assisted (or he just evaluated the obtained data from them). The quality of the figures is sometimes confusing (e.g. fig. 24)and tables are appropriately shown as well. In Section 4 - Results and discussions, the main results are summarized in seven subchapters.

The Results and discussions deal mainly with the "traditional" characterization techniques (such as PSD, XRD, Raman spectroscopy, SEM, EDX, DRS) and the photocatalytic evaluation of the as-obtained materials (in the presence of MB and RhB as model pollutants). The reusability and the antimicrobial assessment of samples are also presented. Some typos are present, but much fever compared to the previous version of the Thesis.

All new scientific results (presented in 5 main points) are defined and acceptable. In addition, the applicability of the scientific results was also proved in Claim 5 *via* successful attempts regarding the antimicrobial properties against *Vibrio fisheri*.

Using the results mentioned above, Engr. Asfandyar Khan has published two journal articles (with a cumulative IF of 8,64). Moreover, he had six conference presentations, including five oral and a poster presentation (mainly from the pre-pandemic period, suggesting that in "normal" conditions, these numbers would be much higher). Thus, the requirements of the doctoral school are plenty fulfilled.

Moreover, to give You the possibility to show your debate skills, I would like to propose some of my main questions here. These are mainly coming from my curiosity and free of challenging doubts regarding the work:

- You have used mainly "relative degradation efficiencies" to characterize the efficiency of the catalysts. Although, in this way, if the selected reference catalyst has inferior performance, then the performance increment can reach more than 1000% (but, in reality, can degrade just e.g. 10% of the contaminant vs. the 1% efficiency of the control). In extreme cases, if the control has an efficiency of ≈0%, the enhancement will be close to infinite. Why have you used this way to characterize these efficiencies instead of the classic definition/presentation of yields?

- Regarding the BET results: you say that the catalyst consisting primarily of spherical and small needle-like structures have a lower surface area compared to the samples with larger needles. Was there any evaluation about the ratio of spherical/needle-like structures of the NP1-3 samples?
- The specific surface area values and the band gap values have a "linear" enhancement in the range of samples NP1-NP6 (the value of SSA increases from NP1 to NP5/6, while the band gap values are decreasing). Although, in term of relative degradation efficiencies (fig 32), the samples NP2-3 were proved to be the best photocatalyst. How can you explain this (apparent) contradiction between these results? (as usually it is said, that the high SSA and low band gap values are benefic for high photocatalytic efficiencies)
- How can you explain that compared to the conventional Fenton systems (which are usually performing better at lower pH), your system showed better performance at high pH values?
- Which is the equation for the trendline shown in Fig. 33? (if there is not a defined equation then you should not use this option)
- How can you detail the mechanism proposed about the attachment of copper-based nanoparticles? How can this behaviour be compared to the literature?/are there any references investigating the antibacterial effect of similar nanomaterials?
- How can you explain the unusual behavior/effect of different concentrations of H_2O_2 on the degradation of MB, presented in Fig. 29?
- In Chapter 4.6, it is observed that the NP-3 has its best performance around 3-4 cycles. How can you explain this (the increase compared to the first cycle and the decrease after the 4^{th} cycle)?

Results summarized in the Thesis can be **accepted;** the work is suitable to be defended in front of the committee and the larger audience. Moreover, if the level of presentation and discussion skills shown during the defence will be comparable with the quality of the written form of the research, I'll give my consent to confer the PhD degree in Engineering Sciences to Mr. Engr. Asfandyar Khan.

Dr. Gábor Kovács