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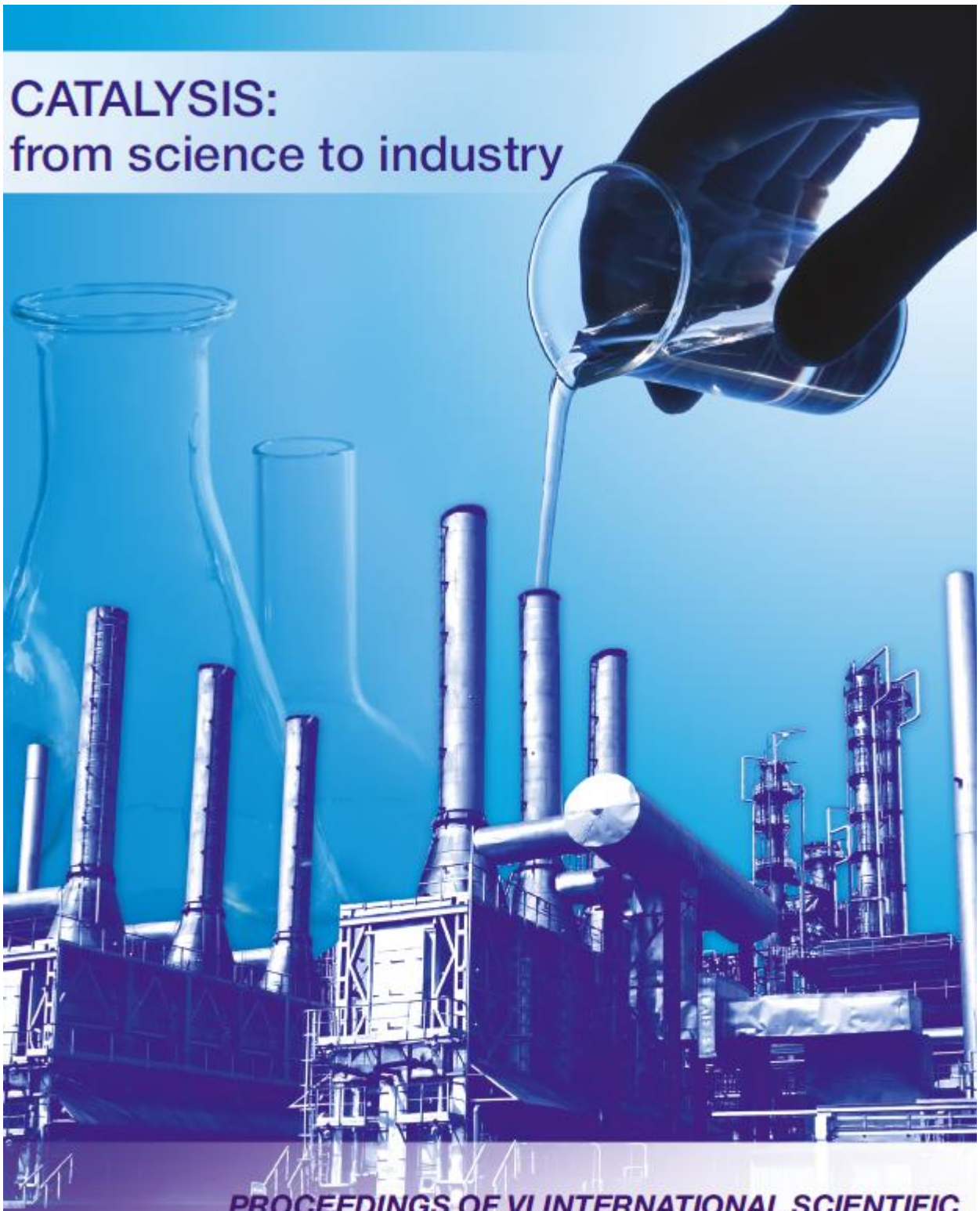

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Design of Ag-CeO_2/SBA-15 catalysts for room-temperature 4-nitrophenol reduction

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Catalytic hydrogenation of nitroaromatic compounds into corresponding aminoaromatic compounds using environmentally safe materials is an attractive approach to address two challenges: efficient removal of the nitroaromatic compounds from wastewaters and enlarging the manufacturing scale of the aminoaromatic compounds employed in pharmaceutical, chemical, and agrichemical industries [1, 2]. The Ag/CeO_2 system is a promising catalytic composition with strong metal-support interaction resulting in charge transfer over the interface and cooperative action of the metal and oxide active sites in catalytic reactions [3]. However, Ag/CeO_2 systems tend to form aggregates of large size, and this substantially decreases the active surface and catalytic activity. It is required to utilize primary support, for instance, SiO_2. The SBA-15 material is a mesoporous silica with hexagonally ordered cylindrical pores and high surface area (550–900 m^2/g) [4] allowing its application as a catalyst support. The work aims to prepare Ag-CeO_2/SBA-15 system and study the synergetic effect of the Ag-CeO_2 interface on catalytic activity in the 4-nitrophenol (4-NP) reduction into 4-aminophenol (4-AP) at room temperature.

The SBA-15 was prepared by the template method [5] using triblock copolymer Pluronic P123 (BAFS, Germany). Silver- and/or ceria-containing catalysts (Ag/SBA-15, CeO_2/SBA-15, Ag-CeO_2/SBA-15) were prepared by the incipient wetness impregnation of the Pluronic P123@SBA-15 hybrid using $AgNO_3$ and/or $Ce(NO_3)_3 \cdot 6H_2O$ aqueous solutions with addition of citric acid. Nominal loading of silver and ceria in the catalysts was 5 and 10 wt.%, respectively. The synthesized samples were characterized by N_2 sorption, XRD, SAXS, UV-vis DRS, H_2-TPR, and tested in 4-NP reduction into 4-AP by sodium borohydride ($NaBH_4$) at room temperature and atmospheric pressure in water medium.

According to results of low-temperature N_2 sorption, the sinthesized samples possess high surface area (594–754 m^2/g), large pore volume (0.86–1.04 cm^3/g), and a narrow pore size distribution in the range between 5.5 and 7.8 nm. The XRD and SAXS data indicate the formation of Ag and CeO_2 particles with sizes up to 10 and 3-4 nm, respectively, and justify hexagonally ordered porous structure of the SBA-15 material. Both the simultaneous reduction of the AgO_x and surface of CeO_2 particles in TPR and the red shift of the surface plasmon resonance (SPR) peak of Ag nanoparticles in UV-vis spectra for Ag-CeO_2/SBA-15 catalyst indicate the presence of Ag-CeO_2 interface in catalysts. The reduction of 4-NP reaction does not proceed without the catalyst. The support (SBA-15) and the CeO_2/SBA-15 sample show rather low activity in the process, while Ag-containing samples (Ag/SBA-15 and Ag-CeO_2/SBA-15) are active, with the latter showing superior activity. Rate constants were calculated within pseudo-first order reaction model and amounted to $k_{exp} = 0.010\ s^{-1}$ and $k_{exp} = 0.016\ s^{-1}$, respectively.

To conclude, the obtained samples possessed the high surface area and narrow pore size distribution. The key role in stabilization of the dispersed Ag and CeO_2 particles (<5 nm) with the developed active surface was attributed to the confined porous structure of the SBA-15 serving as a nanoreactor, application of citric acid, and additional stabilization features of the triblock copolymer. The study of the optical properties and the reduction features indicated the developed Ag–CeO_2 interfacial interaction in the Ag-CeO_2/SBA-15 system that enhanced the catalytic response in the 4-NP reduction.

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