Diffusion Processes in Nanostructured Three-Way Automotive Powder Catalysts

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Catalytic properties of nanosized perovskite $LaCoO_3$ and $LaMnO_3$ powders are considered experimentally and theoretically. For the purpose of the use of those catalysts in automotive exhaust gas conversion the kinetics of oxygen atom transport processes from/to gas phase and catalyst are experimentally analyzed by isotopic oxygen exchange method. Molecular oxygen isotope $^{18}O_2$ gas is introduced into reactor with powder of catalyst. The process of exchange is performed at temperature 400^{o}C. As a result of oxygen exchange between gas as catalyst the molecular species of oxygen $^{18}O_2$, $^{18}O^{16}O$ and $^{16}O_2$ appears in gas phase which kinetics is registered by mass spectrometer. Monocrystalline and polycrystalline powder particles are prepared [1,2] are considered.

The obtained experimental kinetic curves of partial pressures of oxygen species are fitted by proposed real time kinetic model based on rate equations. Model includes processes of chemical reactions (complex and simple heteroexchange) and diffusion of oxygen inside powder nanoparticles. The diffusion process is introduced considering the bulk diffusion adapted for powder catalysts [3] and diffusion in grain boundaries for polycrystalline particles. The main attention is paid on grain boundary diffusion.

Calculated curves are in a good agreement with experimental points for all three types of catalysts. In the case of polycrystalline powder particles both bulk and grain boundary diffusion takes place, while in monocrystalline case only bulk diffusion occurs. This assumption is realized in model. From the calculated results the kinetic (exchange rate constants, diffusion coefficients of bulk and grain boundary diffusion) and thermodynamic (activation energies of exchange reactions and diffusion) parameters are obtained.

References:

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