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Catalysis solves environmental problems of nuclear industry

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Recent advances in application of catalysis to problems of waste destruction and off-gas treatment in nuclear industry will be presented in this paper. The research activities of Boreskov Institute of Catalysis (BIC) in this field proceed in four main directions: (1) destruction of mixed organic wastes in a fluidized catalyst bed, (2) waste oxidation in molten salts, (3) selective catalytic reduction of NO_x in off-gases, (4) catalytic oxidation of H_2 in the ventilation exhaust from liquid waste tanks, (5) catalytic removal of VOC, NH_3 and CCl_4 in groundwater and soil remediation.

One of the most hazardous types of wastes in nuclear industry is so-called mixed wastes containing organic compounds: oils, extractants and solvents contaminated with radionuclides of uranium, plutonium and fission products. These wastes represent a threat to the environment upon their storage and treatment because of the combination of flammability, explosiveness, toxicity and radioactivity. Both in Russia and the USA huge amounts of such wastes are accumulated in tank farms at Hanford and other nuclear sites. For environmentally clean destruction of mixed organic wastes we proposed a new technology based on flameless low temperature oxidation of wastes in a fluidized catalyst bed and transition of radionuclides to compact solid phase for further processing or disposal. The tests revealed the following main advantages of the proposed waste treatment technology: high efficiency of waste destruction; suppression of the formation of pollutants (NO_x , CO, HC); and elimination of secondary waste streams. The technological scheme of the process of mixed waste treatment includes a catalytic reactor for waste destruction and an off-gas treatment system. The experiments in two pilot fluidized bed reactors, carried out both with model systems and real wastes, showed a high efficiency of waste destruction, the clean exhaust does not contain radionuclides and toxic compounds, such as dioxins, nitrogen oxides, sulfur oxides, and CO.

At present a large-scale (50t/year) demonstration plant for the treatment of organic wastes from fuel rods production, containing uranium isotopes is being assembled at Novosibirsk Chemical Concentrates Plant. A new project is started in 1998 for the development of the technology of catalytic destruction of mixed organic wastes of the Siberian Chemical Complex in Tomsk-7, containing oils and extractants, contaminated with enriched uranium (up to 90%) and plutonium.

Another promising technology of mixed waste treatment that meets environmental standards is oxidation in catalytically active molten salts. The destruction of various organic compounds -- methane, propane, chlorobenzene, carbon tetrachloride, polyvinylchloride, and CO -- was carried out in the melts: NaOH--KOH ; LiCl--KCl ; $\text{Li}_2\text{SO}_4\text{--Na}_2\text{SO}_4$; $\text{Li}_2\text{CO}_3\text{--K}_2\text{CO}_3$; carbonates doped with transition metal oxides (Fe_2O_3 , Cr_2O_3 , Co_3O_4 , MnO_2) and catalytically active $\text{K}_2\text{O--V}_2\text{O}_5$ system (pure and doped with CuO). The effects of melt composition, oxygen concentration, load on melt

and temperature on destruction efficiency were studied. Catalyst performance depends on its composition and operational conditions.

Thermal methods of mixed waste treatment, especially plasma arc process, currently used in the USA, produce high levels of NO_x in off-gas. This is largely due to high concentrations of nitrates and nitrites in waste and atmospheric nitrogen being converted to NO_x at high temperatures. Nitrogen oxides are removed most efficiently by Selective Catalytic Reduction (SCR) by ammonia. Conventional commercial SCR catalysts cannot be disposed of in US land fills because of very restrictive land fill disposal regulation. The new bulk and supported monolithic honeycomb catalysts with environmentally benign composition were tested in comparison with a commercial SCR catalyst based on vanadium and tungsten oxides supported on titania. The tests showed that new environmentally benign catalysts provided 85-90% NO_x removal efficiency at space velocity ca. 55 000⁻¹ and temperature 325-375°C. Catalysis can be successfully applied for solving of the safety problem of hydrogen from waste tanks and other apparatuses in the nuclear industry. In the particular case of liquid mixed wastes represented by aqueous solutions containing radionuclides stored in tanks for considerable time, due to water radiolysis and homogeneous catalytic processes hydrogen emission occurs, and explosive gaseous mixtures in the upper part of a tank are formed. Ventilation of tanks to dilute the hydrogen do not guarantee complete safety, because the hydrogen formation is very irregular and it can erupt in large bubbles ("burping" Hanford tank 101 SY). The explosion of hydrogen, which can be initiated by radiation, will lead to severe environmental consequences and radioactive contamination of large territories. The Pt and Pd catalysts on hydrophobic honeycomb structures have been developed and tested in an original apparatus with circulation of hydrogen containing tank gas.