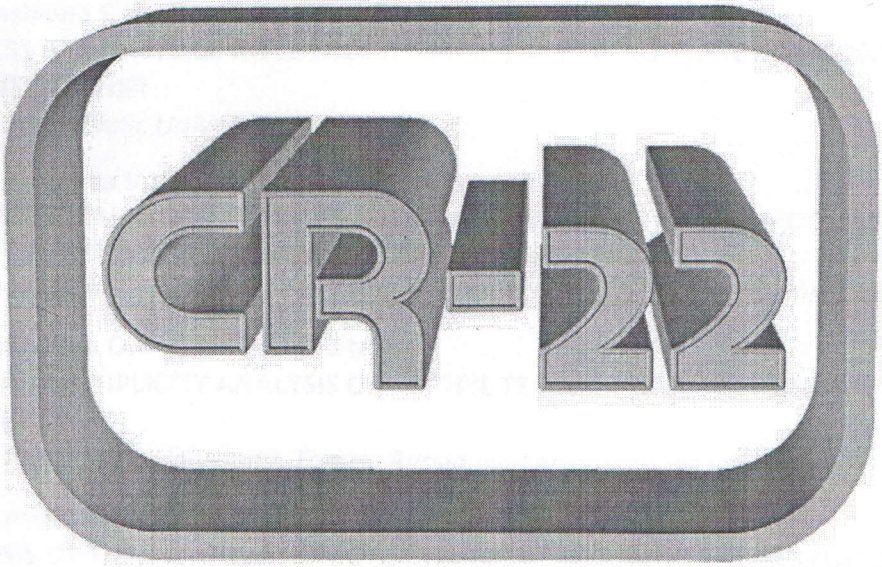

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Silicon-Tungsten Heteropoly Compounds - The Active Catalyst Components of Incomplete Oxidative Conversion of C_1-C_2 Alkanes

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The combination of acid and redox properties is characteristic for heteropoly compounds (HPC). Numerous studies on use of HPC as catalysts of acid and redox type are known. Physico-chemical properties of W-HPC under conditions of the high temperature of catalysis and effect of reaction medium were insufficiently investigated. Therefore, identification of the nature of catalytic action of HPC in high temperature processes including oxidative conversion of hydrocarbons is difficult.

This paper presents experimental data on the study of thermal stability of $H_4SiW_{12}O_{40}{\times}nH_2O$ heteropoly acid (SiW-HPA) and its salts under the action of reaction medium at oxidative conversion of CH_4 or C_2H_6 at temperatures from 20 to 900°C.

The initial structure of SiW-HPA was studied by temperature programmed reduction (TPR) and XRD methods. The nature of oxygen-containing fragments involved in the interaction with H_2 in TPR process by H_2 was determined.

Investigation of influence of steam-processing of HPA on its structural characteristics by IR spectroscopy, TPR, and XRD methods allowed to show that $H_4SiW_{12}O_{40}$ is characterized by 4 types of structural oxygen: W-O-W angled, W-O-W linear, W=O, Si-O-W. They are characterized by a.b. in the IR spectrum 790, 890 cm^{-1} (W-O-W, angled and linear, respectively) 980-1020 cm^{-1} (W=O), 930 cm^{-1} (Si-O-W), in XRD diffraction peaks in the area 2θ = 5-20° (8-9°). 4 types of reactive oxygen species are observed in the TPR spectra. The first two of them belong to the reduction of the HPC structure. Molecular structure of HPA is stable in vapor-air medium in the temperature range of 20-400°C. Changing in the secondary structure is observed at T > 400°C due to dehydration. HPA destruction begins from 550°C

(TPR, IR, and XRD) from gradual destruction of structural fragments of heteropoly anion. However fragmentary formations (Si-O-W, W=O, in the IR spectra - a.b. 930 and 1020 cm^{-1}, respectively), probably are saved in the field of 650-900°C at vapor-air processing. Partial preservation of the absorption bands, which are characteristic for HPA structure in IR spectra points to that. In parallel, with the preservation of structural fragments of the SiW-HPA there is a formation of oxide W phase (VI) of the tetragonal, and then orthorhombic coordination (apparently on the basis of fragments that have undergone destruction) in the presence of O_2 and water vapor at T = 650-900°C.

Thus, it was installed that HPA preserves the secondary structure at the conditions of reaction medium for oxidative conversion of C_1-C_2 alkanes (hydrocarbons, air, water vapor) at 20-400°C. At the temperatures > 650°C the oxide-similar W compounds are formed together with preserving the fragments of HPA structure.

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