

Molecularly imprinted polymeric Cu(II) catalysts with modified active centres mimicking oxidation enzymes.

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Streszczenie

In this paper the preparation of surface molecularly imprinted Cu(II) catalysts with modified active centres is described. The resins were synthesized using suspension polymerization of 1-vinylimidazole, acrylonitrile, 1,1,1-tris(hydroxymethyl)propane trimethacrylate and 4-vinylloxybutylstearate in the presence of Cu(II) ions and 4-methoxybenzyl alcohol as the template. The resins obtained in the form of polymer beads possess diameters ranging from 30 μm to 300 μm and specific surface areas up to 315 m^2/g depending on sonication mode. The resins, after copper removal, were modified in a two-step reaction: bromoalkylation with four alkyl bromides and subsequent exchange of bromide for tetrafluoroborate and trifluoroacetate anions. Non-modified and modified polymers bearing imidazole ligands inside the active centres were complexed with Cu(II) ions, leading to various copper content and applied as heterogeneous catalysts. The formed Cu(II) complexes were subsequently characterized by electron paramagnetic resonance (EPR) spectroscopy. A comparison of the EPR parameters, obtained by computer simulations of the experimental spectra of different Cu²⁺-catalysts allows to propose structures of the Cu(II) active centres as mostly N₂O₂ type (similar to the enzymatic ones). The catalytic activity of the Cu(II) loaded polymers without and with ionic liquids analogues inside active centres was tested using hydrogen peroxide in model oxidation reaction of hydroquinone. Protonation of imidazole nitrogen and introduction of additional free groups possessing strong electrostatic properties resulted in significant increase of catalytic activity in comparison to non-modified samples (oxidation degrees from 50 % to 90 %, selectivity up to 100 %). Trifluoroacetic anion has stronger effect than other studied anions

Słowa kluczowe

Molecularly imprinted polymers, Imprinted catalysts, Oxidation enzymes, Copper-oxidases, Ionic liquids

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