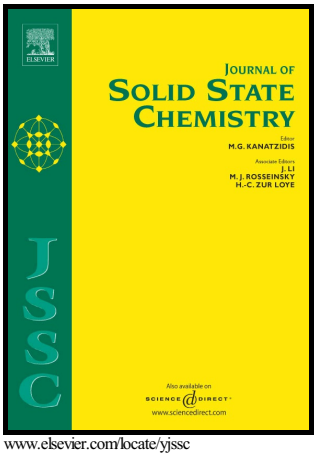

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SYNTHESIS AND STRUCTURE OF PYRIDINE-FUNCTIONALIZED MESOPOROUS SBA-15 ORGANOSILICAS AND THEIR APPLICATION FOR SORPTION OF DICLOFENAC

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Abstract:
A series of pyridine-functionalized mesoporous silicas have been prepared for the first time via direct co-condensation of tetraethoxysilane (TEOS) and 2-(2-pyridyl)ethyltrimethoxysilane (PETS) using the block copolymer Pluronic P123 as a structure-directing agent. The obtained materials were fully characterized by a wide range of instrumental techniques and employed as adsorbents for the removal of a diclofenac which is considered a priority hazardous drug. The synthesized materials exhibit a high adsorption capacities and rapid adsorption rates. The structural and adsorption properties depend largely on the relative amount of PETS/TEOS ratio: the gradual degradation of ordered structure and porosity was observed with the increasing amount of PETS. However due to the highest loading of pyridine units the most structurally degraded material had the highest adsorption uptake (631 mg g^{-1}) indicating that the surface chemistry plays – along with porosity – an important role in governing the adsorption process. The experimental adsorption data were modelled using the Langmuir, Freundlich and Langmuir-Freundlich isotherms – among them the Langmuir-Freundlich model turned out to be the most suitable for describing adsorption behavior of diclofenac onto the materials. The collected data show that the pyridine-functionalized mesoporous silicas can be a promising absorbent of pharmaceuticals.

1. Introduction

Since their discovery in 1992 [1,2] the Ordered Mesoporous Silicas (OMS) have been intensively studied in many laboratories throughout the world, initially due to their outstanding porosity-related properties like high specific surface areas, well-ordered pore structures with adjustable uniform mesopores, and large pore volumes. Tailoring porosity via controllable synthesis protocols and a wide range of accessible templating surfactants is relatively easily achievable. One of the most popular OMS structures is hexagonally ordered SBA-15 (Santa Barbara Amorphous) discovered in 1998 characterized by large mesopores (up to 20 nm), thick pore walls and the presence of irregular interconnecting micropores network [3–5]. SBA-15 materials possess higher thermal and hydrothermal stabilities when compared with MCM-41 materials due to thicker mesopore walls. SBA-15 materials are synthesized under strong acidic conditions in the presence of amphiphilic block copolymers (usually triblock Pluronic P123). The synthesis is carried out via controlled hydrolysis and condensation of tetraethoxysilane (TEOS) followed by removal of the polymeric template by either extraction or calcination. During the first years after discovery a lot of efforts have been devoted to thorough description of the relationships between the synthesis conditions (temperature, time, type of template) and final structure (porosity, ordering, morphology), thus a bunch of excellent papers can be found in the literature [6–11].