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Interference Microscopy Highlights Properties and Peculiarities of SAPO STA-7 Crystals

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1. Abstract

In the framework of this study a new generation of SAPO STA-7 crystals has been investigated with the help of Interference Microscopy. The ability of the above-mentioned technique to record intracrystalline concentration profiles during uptake/release of guest molecules revealed oddities of the system under study. In other words, these crystals have the tendency to break in the middle, enhancing in this way diffusion. On the other hand, molecules have to confront high surface barriers when they try to diffuse through the other sides of the crystal, where it is not broken.

2. Introduction

The need to investigate the sorption properties of porous materials, aiming at the characterization of both the internal structure and surface of these crystals, is more than well known nowadays. Interference Microscopy contributes to this field with great success, due to its unique ability to monitor transient intracrystalline concentration profiles during ad- and desorption [1]. Such profiles are of great importance because they reveal crucial information regarding the uptake processes, such as (i) the direction of internal transport, (ii) the presence of internal structural defects and (iii) of surface/internal barriers.

3. Experimental Section

Interference Microscopy (IFM) is based on the principle that the refractive index of a medium is a function of its composition, which in the present case means that it is a function of the concentration of guest molecules within the crystal. Changes in intracrystalline concentration during uptake/ release of guest molecules by/from the crystal under study will affect the refractive index (with a linear interdependence) and, subsequently, a time dependence of the optical path length of light going through the crystal will be observed. In other words, the changes in the concentration profiles (in particular in the integrals in observation direction) are transferred to changes in the optical path length [2].

4. Results and Discussion

SAPO STA-7 has been found to posses a tetrahedrally connected framework with a three-dimensional interconnected small pore channel system [3]. The functionality and connectivity of its pore system, however, has not yet been explored by IFM.

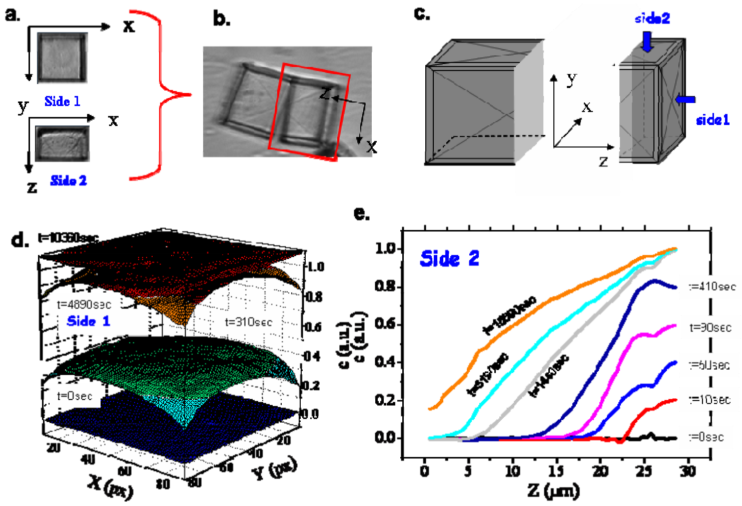

Fig.1: (a) The *xy*- and *xz*-planes of STA-7 30%Si crystal (b) These crystals break after heating at 200^{o}C under vacuum (c) Schematic representation of a broken crystal (d) 2D and (e) 1D intracrystalline profiles during adsorption of methanol

The evolution of the two-dimensional profiles during uptake (Fig.1d) reveals concentration patterns typical of strong surface barriers. This statement is also confirmed by the one-dimensional profiles shown by Fig. 1e. They have been observed after turning the crystal so that now the *y*-axis becomes the direction of observation (side 2). The intracrystalline concentration at big values of *Z* (indicating the central plane where the crystal is broken) rises gradually, whereas at small values of *Z* the surface barriers are so strong that no guest diffusion into the crystal is observed.

In this way, for the first time crystal breaking and the consequences of this phenomenon on the various faces will be directly monitored via Interference Microscopy. It was in particular found that crystal breaking in the centre significally reduces the transport resistance at the crystal surface, though the limitation in surface permeability is still clearly visible in the observed concentration profiles.

References

[1] E. Lehmann et all. , *J. Am. Chem. Soc.,* **2002**, *124*, 8690

[2] U. Schemmert et all. , *Microporous and Mesoporous Materials,* **1999,** *32,* 101–110

[3] P. A. Wright et all., *J. Chem. Soc., Dalton Trans.*, **2000,**1243-1248