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High pressure synthesis of novel, zeolite based nano-composite materials

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Meso/micro-porous solids such as zeolites are complex materials exhibiting an impressive range of applications, including molecular sieve, gas storage, catalysis, electronics and photonics. We used these materials, particularly non catalytic zeolites in an entirely different fashion. In fact, we performed high pressure (0.5-30 GPa) chemical reactions of simple molecules on a sub-nanometer scale in the channels of a pure SiO_2 zeolite, silicalite to obtain unique nano-composite materials with drastically modified physical and chemical properties. Our material investigations are based on a combination of X-ray diffraction and optical spectroscopy techniques in the diamond anvil cell. I will first briefly show how silicalite can be easily filled by simple molecules such as Ar, CO_2 and C_2H_4 among others from the fluid phase at high pressures, and how this efficient filling removes the well known pressure induced amorphization of the silica framework (Haines et al., JACS 2010). I will then present on a silicon carbonate crystalline phase synthesized by reacting silicalite and molecular CO_2 that fills the nano-pores, at 18-26 GPa and 600-980 K; after the synthesis the compound is temperature quenched and it results to be slightly metastable at room conditions (Santoro et al., PNAS 2011). On the other hand, a stable at room condition spectacular crystalline nano-composite is obtained by photo-polymerizing ethylene at 0.5-1.5 GPa under UV (351-364 nm) irradiation in the channels of silicalite (Santoro et al., Nat. Commun,, in press 2013). For this composite we obtained a structure with single polyethylene chains adapting very well to the confining channels, which results in significant increases in bulk modulus and density, and the thermal expansion coefficient changes sign from negative to positive with respect to the original silicalite host. Mechanical properties may thus be tuned by varying the amount of polymerized ethylene. We then think our findings could allow the high pressure, catalyst free synthesis of a unique generation of technological, functional materials based on simple hydrocarbons polymerized in confining meso/micro-porous solids.