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First principles study of the bilayer (alumino)silicate / Ru(0001) heterojunction

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Zeolites are important industrial catalysts with porous structures. The active sites are located inside the pores, which are inaccessible for surface science measurements. A zeolite model system, consisting of two-dimensional (2D) (alumino)silicate bilayer films adsorbed on the Ru(0001) surface, have been synthesized for heterogeneous catalysis studies in zeolite chemistry. Understanding the atomic and electronic structures of the (alumino)silicate / Ru(0001) heterojunction is a fundamental step to unravel the structure-function relationship that is essential to the activities of the catalyst.

We carried out van der Waals density functional theory (vdW-DFT) calculations to study the energy level alignment at the heterojunction, and the noble gas trapping and separation in nano-cages and at the confined interface space. We found that the work function of the Ru substrate can be tuned by surface and interface dipole moments caused by charge rearrangements. The charge rearrangements at the interface is a combined effect of the electron transfer and atomic orbital hybridizations between the O p_z and Ru d_{z^2} states. The energy levels are greatly affected by aluminum concentrations of the bilayer film and the oxidation of the Ru(0001) surface.[1] Combined experimental and DFT studies of the (alumino)silicate/Ru(0001) heterojunction revealed the preferred noble gas atom adsorption sites, adsorption energies and adsorption/desorption energy barriers.[2] The increasing desorption energy barriers of Ar, Kr and Xe in the (alumino)silicate film make it a promising candidate for noble gas separation.

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