# Towards fully-relativistic simulations of the adsorption of super-heavy elements on $\alpha$-SiO$_2$ surfaces *

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While in the last two decades super-heavy elements with $Z \leq 112$ have been studied, the focus of the present work is on the chemical properties of element 114. Our theoretical calculations have been motivated by two conflicting gas-chromatography experiments, which aimed on studying the interaction strength of element 114 with a gold surface. The experiment by Eichler *et al.* [1] reported adsorption in the chromatography column at only very low temperatures of approximately $-90°$C, from which they concluded a weak interaction between element 114 and the gold surface. In contrast, experiments performed at GSI [2] observed adsorption at room temperature, indicating a much stronger bond between element 114 and gold. To resolve this conflict further experiments will be performed at GSI within the next two years, where besides gold SiO$_2$ will be used as detector material.

Besides the previously mentioned experiments, extensive theoretical studies on the adsorption of element 114 on gold surfaces were performed using fully-relativistic DFT methods [3], while the adsorption on inert surfaces such as SiO$_2$ were estimated using semi-empirical methods in conjunction with computed properties of atoms, dimers, or small molecules [3, 4]. Unfortunately a fully-relativistic treatment of the entire adsorption process is beyond the capacities of nowadays computing ressources. However, this work can be divided into two steps: (i) extensive studies on SiO$_2$ bulk and surface properties (e.g. stable bulk-phases or possible surface structures and terminations) using a non-relativistic approach; (ii) fully-relativistic calculations on the adsorption process of element 114 on SiO$_2$, where the most stable and interesting surface structures obtained in the first step serve as basis.

So far we have focused on the first task, understanding the surface structure of SiO$_2$ under realistic experimental conditions. For these calculations, the CASTEP code [6] with Vanderbilt-type ultrasoft pseudopotentials [7] and the PBE exchange–correlation functional has been used. The obtained DFT-energies were then used in conjunction with the *ab initio* atomistic thermodynamics approach [5] to evaluate the surface phase diagrams, providing information of the surface stability as function of surrounding temperature and pressure. Starting with bulk systems, our calculations show that at experimental conditions ($p_{O_2} = 10^{-13}$ atm, 100 K$<T<$320 K) the most stable bulk structure is the so called $\alpha$-quartz. Using this crystal structure as basis, various surface orientations and morphologies were studied. We find that the thermodynamically preferred structure is the $\alpha$-SiO$_2$(001) surface, which could assume three different morphologies: one Si-terminated and two different O-terminated structures.

Figure 1 shows the surface free energy, $\gamma(p, T)$, for the most stable surface structures as function of the oxygen chemical potential. Structure **a**, which is Si-terminated, is favored at $\Delta\mu_O < -6.24$ eV ($T \gg 1000$ K under UHV conditions). In the temperature range until ~100 K the surface is O-terminated (structure **b**). The phase diagram shows the existence of a fourth, but less stable structure (structure **d**), which is terminated by a single O-layer.

After understanding the surface morphology of SiO$_2$ under experimental conditions, the next step will be to perform fully-relativistic DFT calculations on the actual adsorption process of elements 112 and 114 and their homologs on a $\alpha$-SiO$_2$(001) surface (structure **b**).

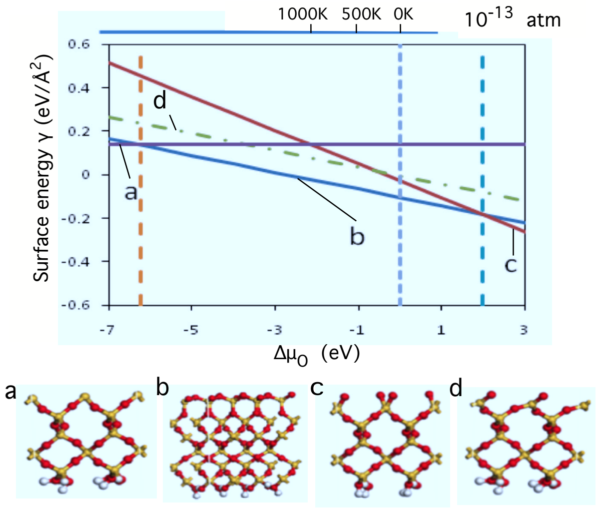

Figure 1: Surface free energy of the $\alpha$-SiO$_2$(001) as function of $\Delta\mu_O$ or temperature at fixed pressure of $10^{-13}$ atm.

## References

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