Goldschmidt 2021 Abstract
https://doi.org/10.7185/gold2021.6830

Phase diagrams of gold-sulfur nanoclusters using atomistic simulations: shape, size and temperature effects

KONO LEMKE

HKU

Presenting Author: kono@hku.hk

We have studied the thermodynamic stability of gold clusters ranging from Au_3 to Au_{17} and their sulfur/hydrogen-containing forms at elevated temperatures using density functional theory and CPMD atomistic simulations. The focus of this work is to construct reliable phase diagrams of $Au_mS_nH_x$ nanoclusters, and to predict their size, composition, shape and distribution in volcanic and hydrothermal ore vapors. We will initially focus on the $Au_3S_nH_x$ and $Au_{17}S_nH_x$ systems, the phase relationships in $Au_3S_nH_x$ and $Au_{17}S_nH_x$, and examine how variations in temperature and pressure influence the stability of $Au_mS_nH_x$. The figure below shows a calculated phase diagram obtained for the gold trimer Au_3 at 298 K under a broad range of partial pressures of H_2 and sulfur gas, together with H_2S isobars at 10^{-5}, 10^{-2} and 10 atm. As seen, in the region of low pH_2 and pS_2 pure Au_3 remains stable, however, when the pressure of H_2 and S_2 increases, adsorption of H_2 and S_2 onto Au_3 edges takes place, producing Au_3H_6, Au_3S_6 and $Au_3S_4H_2$ clusters. Theoretical calculations have also been undertaken to obtain p-T phase diagrams of Au_3S_n and $Au_3S_nH_x$, and these show that Au_3 and Au_3S exist only in low pS_2 vapor and at high temperatures (>800 K), whereas S-rich phases Au_3S_2 and Au_3S_4 become more stable at T≈450-700 K and pS_2≈10^{-7}-10^{-2} atm. This approach has been applied to larger gold clusters, in particular, we have examined interactions between H_2, H_2S and S_n species at edge-, corner- and face-sites of larger Au_{17} cluster, and will present phase diagrams for the $Au_{17}S_nH_x$ system at p-T conditions characteristic of volcanic gases.

