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DFT simulation of the occurrences and correlation of gold and arsenic in pyrite

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Abstract

Using density functional theory (DFT) calculations, the occurrences and correlation of gold (Au) and arsenic (As) in pyrite were studied, and the effect of As on the structural stability of Au in pyrite (FeS_2) was investigated. The calculated results show that Fe is not likely to be simply substituted with Au under normal circumstances. The presence of As is very conducive to incorporating Au into the pyrite interstitial lattice site along with substitution for the S site. It is predicted that a positive correlation exists between Au and As in pyrite and that higher As concentration facilitates the incorporation of Au in pyrite. Additionally, with increasing As content, Au sits on the Fe site and the Fe is repelled into the hole. The pyrite lattice expands with the incorporation of Au and As. Antibonding interactions are found between the Au, Fe, and As atoms. The reducibility of pyrite is greatly enhanced due to the presence of Au and As. The electronic structure calculations show that substituting Au and As for S atoms does not change the pyrite p-type property and that defect energy levels are present in the conduction band. However, with increasing As concentration, incorporating the interstitial site of Au causes a change from the p-type pyrite to an n-type pyrite, and defect energy levels are mainly located in the energy band gap. The interstitial site of Au causes the pyrite to be spin-polarized at a certain As content. In addition, strong interactions are found between Fe 3d and Au 6p orbitals and between Au 5d and As 4p orbitals.

Keywords: Pyrite, gold-arsenic, correlation, DFT calculation