

Density functional theory calculations were employed to investigate the (001), (210), (111), and (110) surfaces of FeS₂. The surface free energies were calculated in equilibrium with a sulfur environment using first-principles based thermodynamics approach. Surfaces that feature metal atoms in their outermost layer are predicted to be higher in energy. Within the studied subset of (111) terminations, the stoichiometric (001) surface terminated by a layer of sulfur atoms is the most stable for sulfur-lean condition. For increasingly sulfur-rich environment, two structures were found to have notably lower surface energies compared to others. They have (210) and (111) orientation, both terminated by layers of sulfur. Interestingly, these surfaces are nonstoichiometric exhibiting an excess of sulfur atoms.