Beyond regular polyhedra: evolutionary prediction and experimental high-pressure synthesis of SiOS

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SiOS represents a ternary generalization of the well-known and highly important family of binary AB_2 compounds including CO_2, SiO_2, GeO_2, CS_2, SiS_2, etc. The presence of two different bond lengths suggests that its crystal structures may be different from those found in SiO_2 and SiS_2. We applied evolutionary search based on DFT *ab initio* calculations to determine crystal structures of SiOS for pressures up to 100 GPa. We predicted the SiOS phase diagram at zero temperature and examined the structural, electronic and vibrational properties of the stable phases. At low pressure the stable phase is a tetrahedrally coordinated layered orthorhombic $Cmc2_1$ structure. This is predicted to transform at 16 GPa to one of two phases with only marginal enthalpy difference: an octahedrally coordinated layered monoclinic *C2/m* structure similar to the *P-3m1* phase of SiS_2 or non-layered octahedrally coordinated tetragonal $P4_2/mmc$. The system remains insulating up to 100 GPa with band gap above 1.4 eV. Following the theoretical prediction we synthesized SiOS by laser heating elemental Si, O and S in the diamond anvil cell at pressure of 8 GPa. The observed XRD pattern is in very good agreement with the theoretical prediction for the $Cmc2_1$ structure.

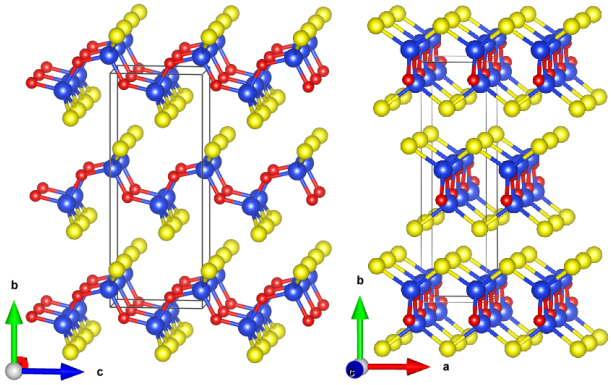

Figure 1: Layered tetrahedral $Cmc2_1$ structure of SiOS stable at low pressures

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