Multipole-Refined Charge Density Study of Diopside

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The electron density distribution of diopside at ambient conditions has been determined by X-ray diffraction and refined through the multipole model of electron density, following the formalism of Stewart [1].

The final multipole refinement includes two extinction parameters, according to the Sabine model, and two kappa expansion/contraction parameters (one for O and the other for Si, Ca and Mg valence monopoles). The expansions over the spherical harmonics were truncated at the octopole level.

Topological analyses of the electron density and its Laplacian distributions [2] reveal that the bonding character of the four Si–O bonds is intermediate between ionic and covalent, as previously found for stishovite and coesite. As expected, the Mg–O and Ca–O bonds are definitely ionic. Determination of the atomic basins results in charges of +3.2(2) *e* for Si and –1.6(1) *e*, on average, for the oxygen atoms. The charges calculated for Mg and Ca are very close to the formal values. Our atomic charges are then more ionic than the corresponding ones determined by Sasaki et al. [3].

The program VALTOPO [4] was used for refinements and topological analysis of the experimental density.

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