Electron density distribution in stishovite, SiO$_2$: a high energy synchrotron radiation study. A. Kirfel$^1$, T. Lippmann$^2$. $^1$Mineralogisch-Petrologisches Institut, Universität Bonn, Poppelsdorfer Schloss, D-53115 Bonn, $^2$GKSS, Max-Planck-Str., D-21502 Geesthacht

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The electron density distribution of the high-pressure polymorph of SiO$_2$, stishovite (a = 4.1773, c = 2.6655 Å, S.G. P4$_2$/mmn, Z = 2), has been redetermined by single-crystal diffractometry using synchrotron radiation of 100.42 keV in order to obtain absorption and extinction free data. The room temperature diffraction experiments on a sample of irregular shape obtained from [1] were performed on the Triple-Crystal Diffractometer installed at beamline BW5 at HASY-LAB/DESY, Hamburg [2]. For a full sphere up to sin$\Theta$/$\lambda$ = 1.35 Å$^{-1}$, 3795 reflections were recorded resulting in a set of 269 unique reflections with an internal agreement factor $R(F^2) = 0.0117$. The structure refinements using the VALRAY programme package [3] converged at $R(F) = 0.0111$, $wR(F) = 0.0131$, GoF = 2.45 for the IAM and at $R(F) = 0.0055$, $wR(F) = 0.0044$, GoF = 0.90 for a multipole model featuring neutral atoms and multipole expansions up to hexadecapoles. For each atom, the radial expansion coefficients of the multipole orders (l>0) were constrained to a common value. Absence of extinction was indicated by a refined correction parameter equalling zero within error limit. The excellent quality of the data is also illustrated by a HO-refinement (s > 0.7 Å$^{-1}$) yielding $R(F) = 0.0068$, $wR(F) = 0.0053$, GoF = 0.85. Both static deformation densities and structure amplitudes compare well with corresponding results from LAPW calculations [4].

Ensuing topological analysis of the total model electron density distribution revealed bond critical point properties for the two unique Si-O bonds, Si-O$_{eq}$ (d = 1.757 Å, $\rho_c = 0.74$ eÅ$^{-3}$, $\nabla^2\rho_c = 11.0$ eÅ$^{-5}$) and Si-O$_{ap}$ (d = 1.808 Å, $\rho_c = 0.62$ eÅ$^{-3}$, $\nabla^2\rho_c = 8.5$ eÅ$^{-5}$). Calculations of the atomic basins and related properties [4] resulted in charges of +3.45 e and −1.72 e for Si and O, respectively. The volumes of the respective basins are 2.32 and 10.48 Å$^3$, corresponding to spheres with radii of 0.82 and 1.36 Å.

The comparison with the results derived from earlier X-ray tube data [5] combining single-crystal and powder intensities shows also reasonable agreement.