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The effect of radiation damage on local structure in the crystalline fraction of $ZrSiO_4$: Investigating the ^{29}Si NMR response to pressure in zircon and reidite

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ABSTRACT

Periodic density functional theory (DFT) was used to calculate the structure of zircon ($ZrSiO_4$) and reidite (scheelite polymorph of $ZrSiO_4$) at pressures from 0-41 GPa. Subsequently, the ^{29}Si nuclear magnetic resonance (NMR) chemical shifts at each pressure were determined using the GIPAW method. The results show that in both zircon and reidite the chemical shifts become more negative with increasing pressure. The chemical shift of reidite is predicted by calculation to be –91.8 ppm and measured as –91.1 ppm. The results are used to interpret the previously observed systematic decrease in ^{29}Si NMR chemical shifts (increasingly negative) in the crystalline fraction of radiation-damaged zircon with increasing α-dose. The original hypothesis that strain induced by the compression of the crystal by internal regions of damaged, amorphized material at high pressure was the origin of the shifts was disproved. An alternative source of internal pressure in the radiation-damaged crystals linked to "'stuffing" of the lattice with interstitial O or He atoms is proposed.