Supplementary Information

Machine Learning-Based XANES Analysis for Predicting Local Structure and Valence in Amorphous Silicon Suboxide

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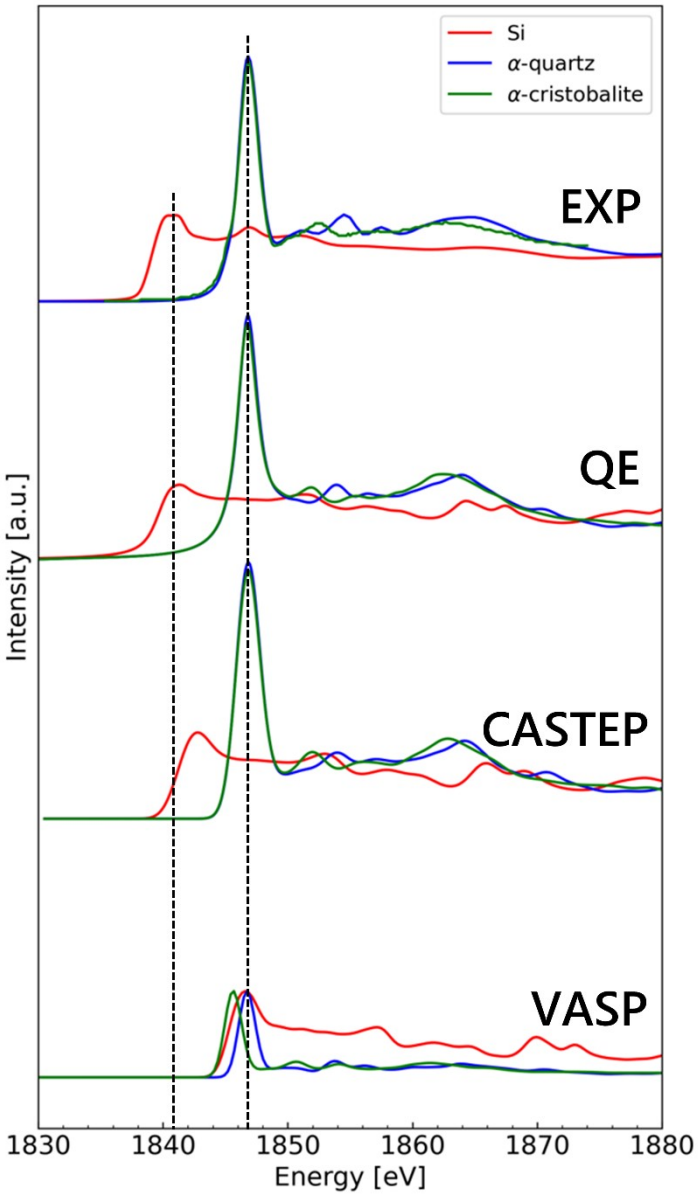

Figure S1. Si K-edge XANES spectra of metallic Si (red), α-quartz (blue), and α-cristobalite (green). The experimental spectrum (EXP) is shown at the top, followed downward by spectra calculated using Quantum Espresso (QE), CASTEP and VASP. The experimental spectra were adapted with permission from Refs. [1], [2], and [3] for metallic Si, α-quartz, and α-cristobalite, respectively.

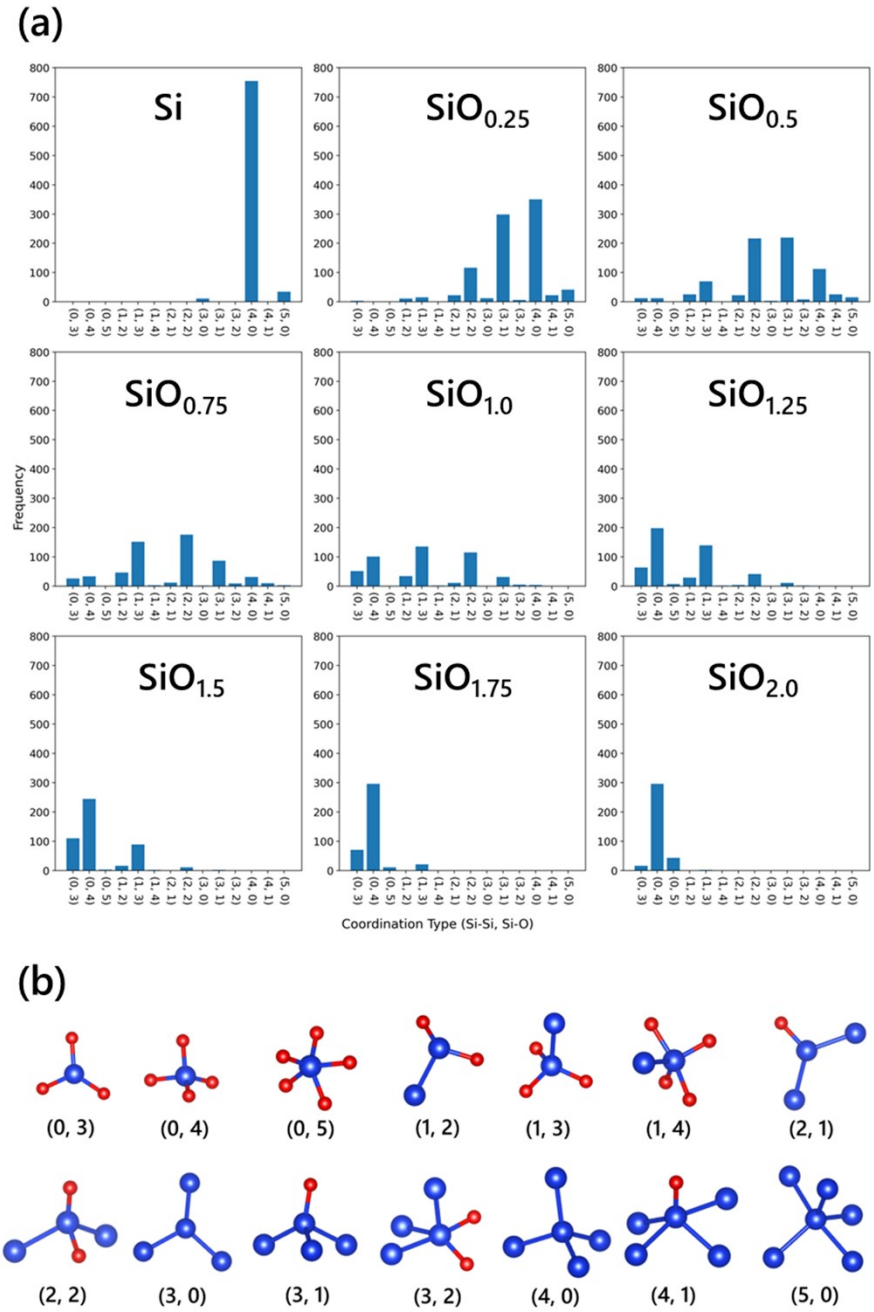

Figure S2. (a) Histograms of the Si–Si/Si–O coordination types in the amorphous SiOx models for nine compositions (x = 0–2, Δx = 0.25). Each panel displays the frequency of the structural units, which are labeled according to the numbers of Si and O atoms bonded to a central Si atom. (b) Representative local structural motifs for each (m, n) coordination environment, wherein the Si and O atoms are represented by blue and red spheres, respectively.

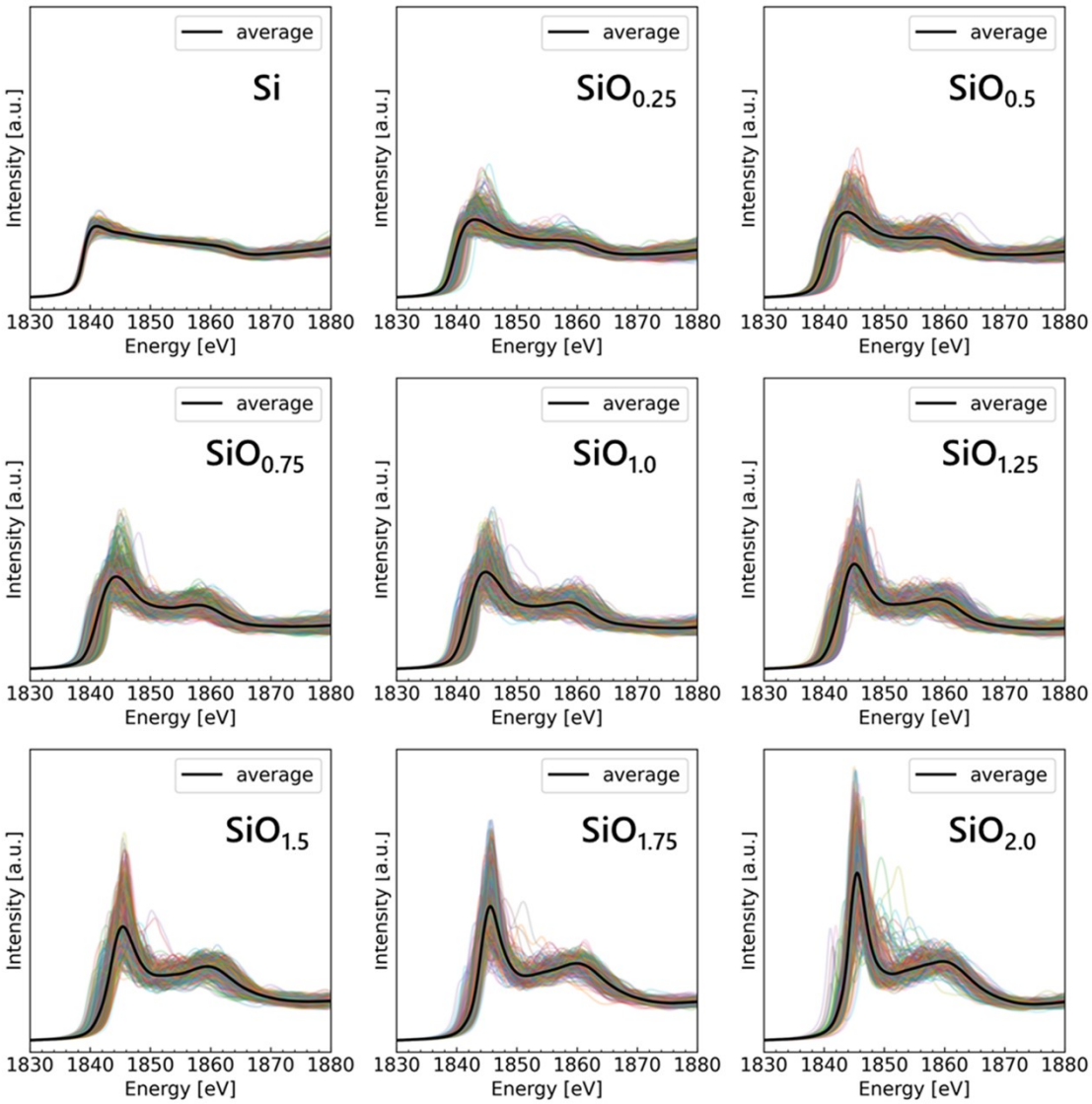

Figure S3. Calculated Si K-edge XANES spectra for the amorphous SiO_x (x = 0–2, step = 0.25). For each composition, the spectra for all individual Si sites are overlaid (colored curves), and the black line indicates the composition-averaged spectrum.

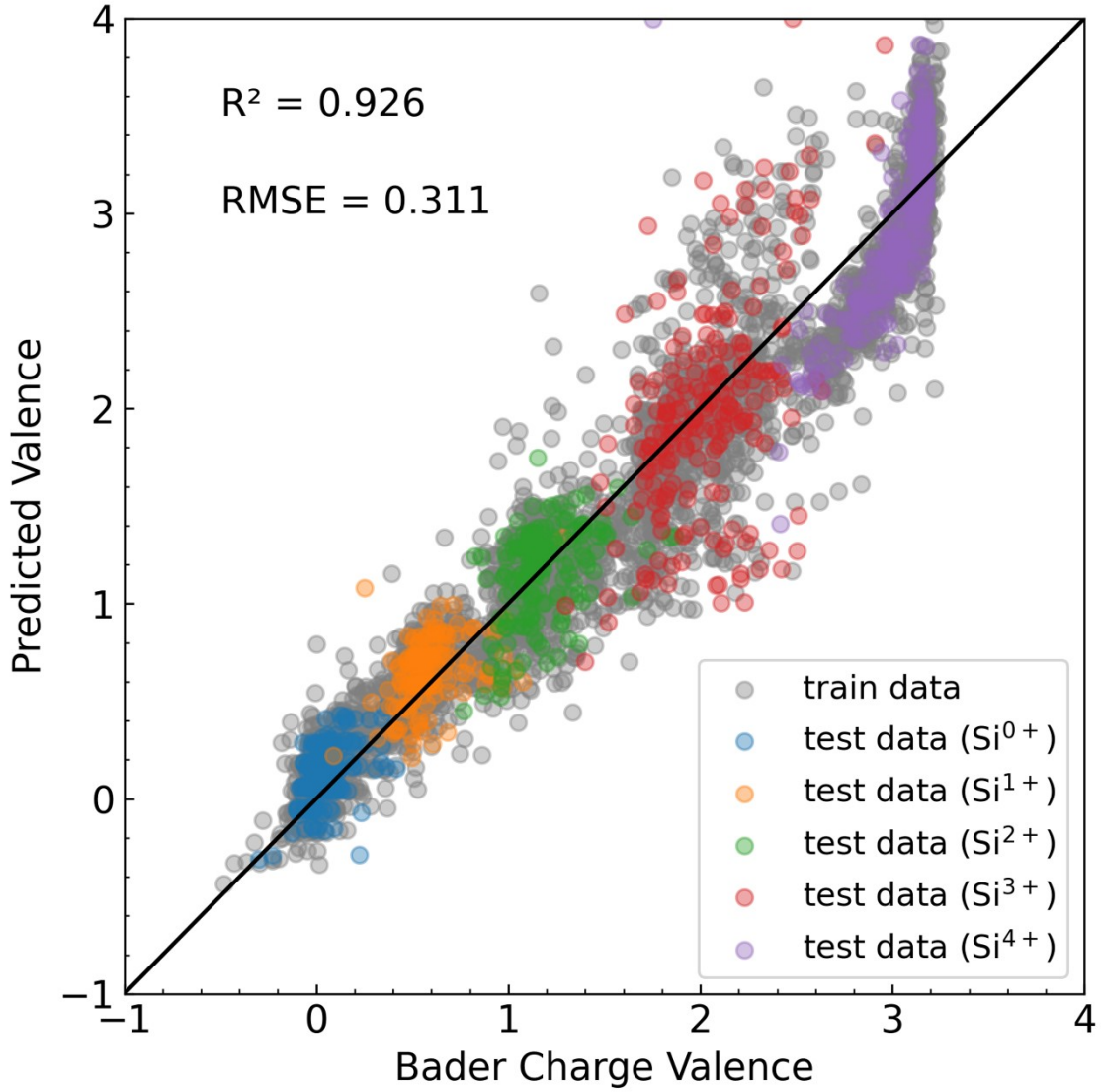

Figure S4. Parity plot of the Si valence predicted by a linear regression model that uses three spectral descriptors: the edge energy, the white-line peak height, and the integrated white-line intensity. Test atoms are color-coded according to their formal valence states: blue, Si^{0+}; orange, Si^{1+}; green, Si^{2+}; red, Si^{3+}; and purple, Si^{4+}. Each valence category corresponds to the number of O neighbors in the first coordination shell (0–4).

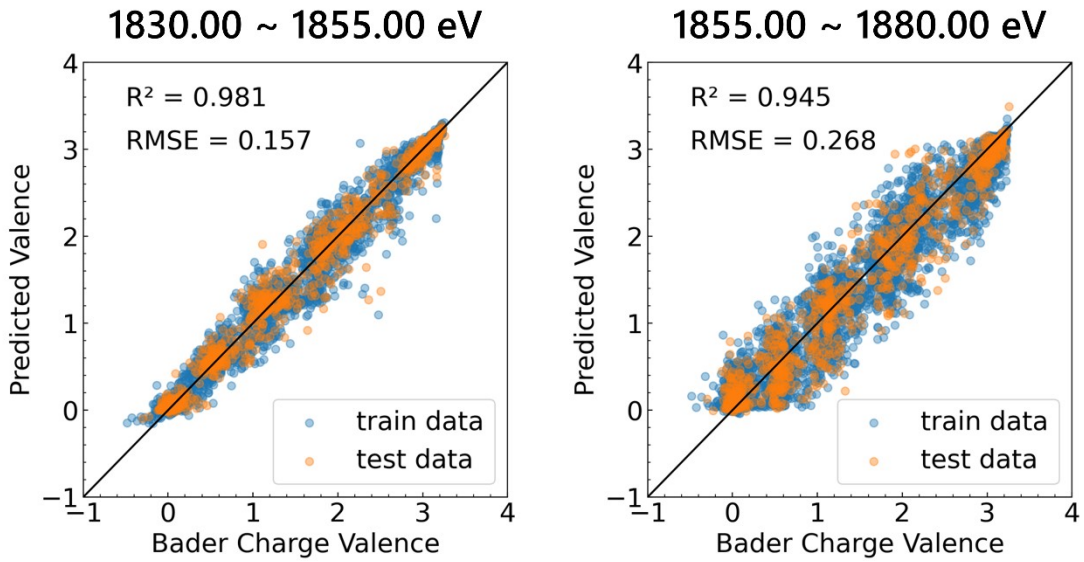

Figure S5. Scatter plots comparing the neural-network-predicted Si valences with the Bader-charge references for models trained on 25 eV sub-windows of the Si K-edge XANES spectrum (the corresponding energy range is indicated in each panel). Blue and orange markers denote the training and test data, respectively, and the diagonal line represents a perfect agreement. The R^2 and RMSE values are annotated in each panel.

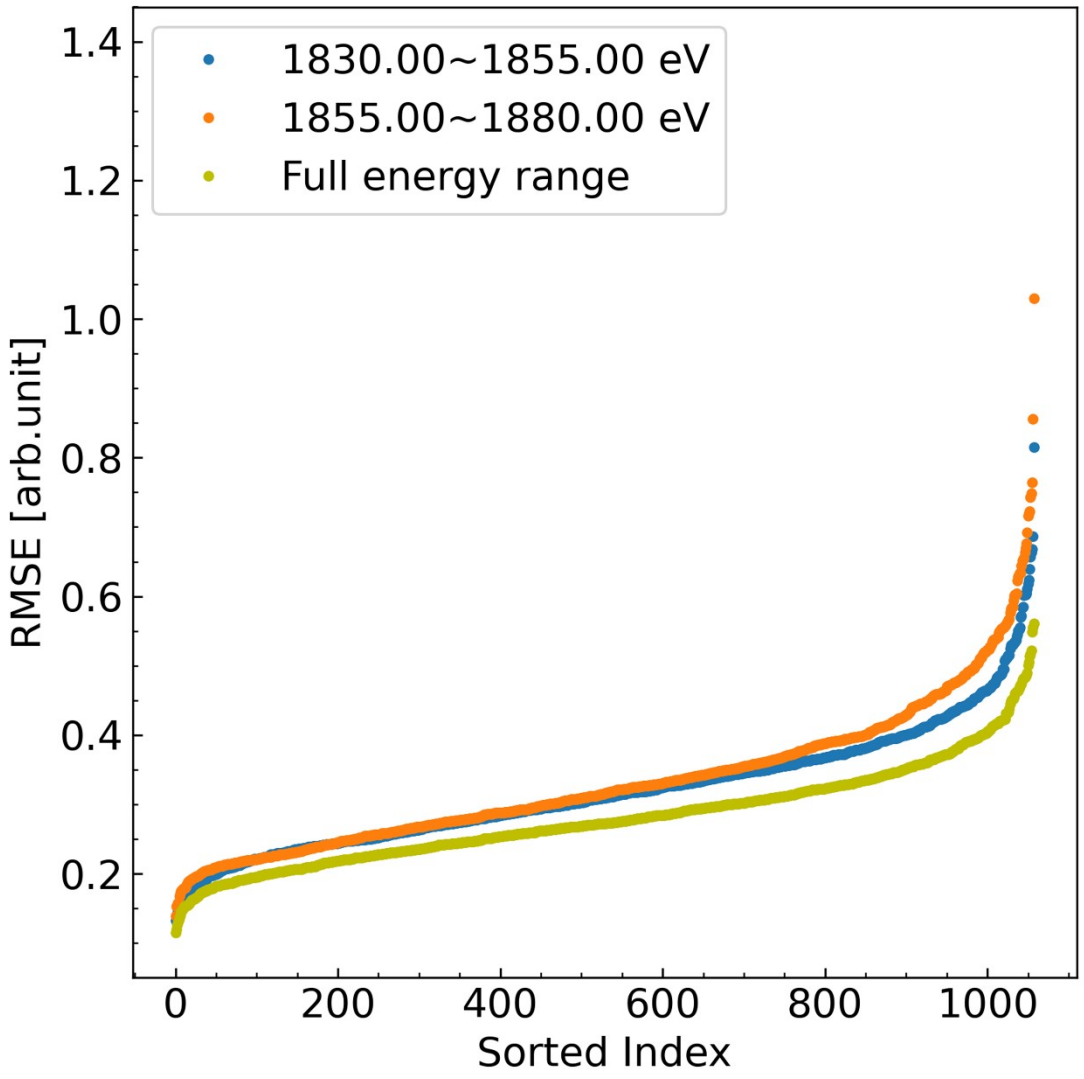

Figure S6. Sorted RMSE distributions for the RDF predictions generated by the neural-network models trained on individual 25 eV sub-windows of the Si K-edge XANES spectrum (orange and blue curves), and by a model based on the full 1830–1880 eV range (yellow curve).

References

1 Masashi Ishii, Ritsumeikan SR Center, XAFS spectrum of Silicon., https://mdr.nims.go.jp/datasets/8aa129b6-8c2c-42a6-ac61-24893890bdc2

2 Masashi Ishii, Ritsumeikan SR Center, XAFS spectrum of Silicon dioxide (Quartz)., https://mdr.nims.go.jp/datasets/c665369d-28b1-4c1d-9198-00f61555fe34

3 D. Li, G. Bancroft, M. Kasrai, M. Fleet, R. Secco, X. Feng, K. Tan and B. Yang, X-ray absorption spectroscopy of silicon dioxide (SiO2) polymorphs : the structural characterization of opal, *American Mineralogist*.