SUPPORTING INFORMATION

Predicting Composition-Structure Relations in Alkali Borosilicate Glasses using Statistical Mechanics

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**Table S1.** Compositions and fictive temperatures (*T*f) of MD simulated potassium borosilicate glasses.

|  |  |  |  |
| --- | --- | --- | --- |
| [K2O]  (mol%) | [B2O3] (mol%) | [SiO2] (mol%) | *T*f (K) |
| 14.3 | 28.6 | 57.1 | 1945 |
| 25 | 25 | 50 | 1829 |
| 40 | 20 | 40 | 1002 |
| 53.9 | 15.4 | 30.8 | 582 |
| 15.3 | 17.0 | 67.8 | 2129 |
| 28.6 | 14.3 | 57.1 | 1666 |
| 37.5 | 12.5 | 50 | 1170 |
| 50 | 10 | 40 | 656 |
| 11.4 | 12.7 | 76.0 | 2452 |
| 22.2 | 11.1 | 66.7 | 2156 |
| 36.4 | 9.1 | 54.5 | 1374 |
| 51.7 | 6.9 | 41.4 | 597 |

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| --- | --- | --- | --- | --- |
|  |  |  |  |  |

**Table S2.** Glass transition temperature (*T*g) of sodium and potassium borate and silicate glasses from literature used to predict the enthalpy values in Table 1 (Belova et al., 2015; Schroeder et al., 1973; Shelby, 1983).

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| [Na2O]  (mol%) | [K2O]  (mol%) | [B2O3] (mol%) | [SiO2] (mol%) | *T*g (K) |
| 15 |  |  | 85 | 770 |
| 17 |  |  | 83 | 780 |
| 19 |  |  | 81 | 773 |
| 20 |  |  | 80 | 770 |
| 22 |  |  | 78 | 767 |
| 25 |  |  | 75 | 764 |
| 27 |  |  | 73 | 758 |
| 29 |  |  | 71 | 754 |
| 30 |  |  | 70 | 723 |
|  | 8 |  | 92 | 820 |
|  | 10 |  | 90 | 781 |
|  | 15 |  | 85 | 758 |
|  | 20 |  | 80 | 743 |
|  | 25 |  | 75 | 732 |
|  | 33 |  | 67 | 700 |
|  | 40 |  | 60 | 679 |
| 1 |  | 99 |  | 532 |
| 10 |  | 90 |  | 628 |
| 15 |  | 85 |  | 678 |
| 20 |  | 80 |  | 723 |
| 25 |  | 75 |  | 743 |
| 30 |  | 70 |  | 746 |
| 35 |  | 65 |  | 729 |
| 70 |  | 30 |  | 514 |
|  | 5 | 95 |  | 571 |
|  | 10 | 90 |  | 616 |
|  | 15 | 85 |  | 657 |
|  | 20 | 80 |  | 693 |
|  | 25 | 75 |  | 715 |
|  | 30 | 70 |  | 717 |
|  | 35 | 65 |  | 700 |
|  |  |  |  |  |

**Figure S1.** Composition dependence of the fraction of *B4* structural units in (a) sodium borate (Schuch et al., 2011; Shelby, 1983) and (b) potassium borate (Zhong and Bray, 1989) glasses. The closed symbols represent 11B MAS NMR experimental data and the solid lines represent the model predictions.





**Figure S2.** Composition dependence of *Qn* fraction in sodium borosilicate obtained by 29Si MAS NMR (Bhasin et al., 1998) (symbols) compared to model predictions (plotted). Results are shown for glasses with (a) *K* = 4 and (b) *K* = 6.





**Figure S3.** Sodium borosilicate structural data obtained by MD simulations (Deng and Du, 2018) compared to model predictions. (a) and (b) *Qn* fractions for silicate structural groups (symbols) and model predictions (lines) plotted against the overall modifier concentration for glasses with *K* = 4 and 6, respectively. (c) and (d) Fraction of four-fold coordinated boron as obtained by MD simulations (symbols) and the model predictions (line) against the overall modifier concentration for glasses with *K* = 4 and 6, respectively.









**Figure S4.** Potassium borosilicate structural data obtained by MD simulations compared to model predictions with zero free parameters. (a) and (b) *Qn* fractions for silicate structural groups (symbols) and model predictions (lines) plotted against the overall modifier concentration for glasses with *K* = 4 and 6, respectively. (c) and (d) Fraction of four-fold coordinated boron as obtained by MD simulations (symbols) and the model predictions (line) against the overall modifier concentration for glasses with *K* = 4 and 6, respectively.









**Figure S5.** Fictive temperatures (Tf) as found by MD simulation, using the method reported by Liu et al. (Liu et al., 2018) and as predicted by extrapolating experimental Tg values (Grandjean et al., 2008) by a constant. We see a significant difference in absolute values, where the simulated fictive temperatures fluctuate from 500 K to 2500 K. Even with the high fluctuation of absolute values, they correlate well with the experimentally obtained *T*f values.



**Supporting References**

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