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# Elaboration of entropy with glass composition: A molecular dynamics study



# Z. Mollaei<sup>a</sup>, F. Kermani<sup>a</sup>, F. Moosavi<sup>b,\*</sup>, M. Kahani<sup>c</sup>, S. Mollazadeh<sup>a</sup>, J. Vahdati Khaki<sup>a,\*</sup>

<sup>a</sup> Department of Materials Engineering, Faculty of Engineering, Ferdowsi University of Mashhad (FUM), Mashhad 9177948564, Iran

<sup>b</sup> Department of Chemistry, Faculty of Science, Ferdowsi University of Mashhad, Mashhad 9177948974, Iran

<sup>c</sup> Department of Computer Engineering, Faculty of Engineering, Ferdowsi University of Mashhad, Mashhad 9177948564, Iran

#### ARTICLE INFO

#### ABSTRACT

Keywords: Entropy Glass composition Glass properties Glass transition temperature Molecular dynamics simulation Recently, entropy was proposed as a simple input into glass science and engineering, which has an interesting relationship with the glass properties containing glass transition temperature (Tg), melting point, and concentration of non-bridging oxygens (NBOs). In the current study, molecular dynamics (MD) simulation as the powerful method was used to approve the recently observed relations. In this regard, various silicate-based compositions containing 25, 30, and 35 mol% of alkaline earth oxides were simulated. The Tg, bond length, and the concentration of NBOs were evaluated using MD simulation results, including volume-temperature curves and radial distribution functions (RDF) results. According to the results, Tg values of the simulated glass were reduced up to 400 K by increasing the amounts of additives up to 35 mol%. The distance between Si and O species as the glass former basis increased from 1.580, 1.591, 1.608, and 1.615 Å to 1.590, 1.596, 1.616, and 1.621 Å, in the SsiO<sub>2</sub>-MgO, SiO<sub>2</sub>-SrO, SiO<sub>2</sub>-CaO, SiO<sub>2</sub>-SrO, and SiO<sub>2</sub>-BaO systems, respectively. Besides, as found from the results, increasing the concentration of additives increases the mobility of Si-O paired atoms. Accordingly, the glass disorderliness or entropy of the systems increases with adding glass modifiers. Also, the concentration of NBOs in the mentioned systems was 35.9-56.3, 36.8-57.5, 26.1-43.9, and 18.2-29.7%. The results of NBOs confirm that the glass disordering increases by increasing amounts of additives. In summary, using MD simulation of silicate glass containing alkaline earth oxides, the relation between entropy as a theoretically calculated property and glass characteristics such as Tg is successfully approved.

## 1. Introduction

Systematic and atomistic studies of materials are among the utmost issues in evaluating, designing, and improving materials [1–3]. For example, configurational entropy ( $S_{conf}$ ),  $S_{Conf} = -R\sum_{i=1}^{x} x_i ln x_i$ , or a parameter to measure disorder in a thermodynamic system, is one of the essential microscopic variables of materials [4]. Transition from macroscopic to the microscopic world is challenging due to the different scales of time and place [5]. The microscopic world contains a huge sea of data, concepts, and information [6]. Molecular dynamics (MD) simulation has evolved into a modern method that sheds light on the atomistic points of view in various materials, from simple to biological ones [7–9]. MD could contribute to investigating deeply relationships between the structure and the function of materials that are usually unattainable from experimental techniques containing X-ray/neutron diffraction, infrared (IR), Raman, X-ray scattering (XAS), and nuclear magnetic resonance (NMR) spectroscopies [10,11].

Recently, configurational entropy was introduced as a microscopic

parameter for evaluating and developing glass materials, i.e., the nonequilibrium and non-crystalline solid products can exhibit the amount of glass transition temperature (Tg) [12]. In addition, it is contributed to drawing correct relation between the entropy, chemical composition, and other properties of glass. Fortunately, MD simulation is applied to approve the contributions between these parameters; it can provide an excellent opportunity to understand the relationship between glass properties and compositions [7,13,14]. Using MD results, structural and dynamical properties of glass containing structure, Tg, and bond length have been studied [15-17]. As reported in [18,19], a distribution of bonds should be studied to find the symmetry change containing translation-rotation symmetry and topological disorder. Furthermore, adding additives to a glass network changes the atom distributions and disorders [20]. In this scenario, MD enables the direct determination of the structure-composition relationship with the possibility of identifying new structures of glass materials.

Recently, it was shown that  $T_g$ , melting point, and the concentration of non-bridging oxygens (NBOs) have a clear relationship with  $S_{conf}$ 

\* Corresponding authors. *E-mail addresses:* Moosavibaigi@um.ac.ir (F. Moosavi), Vahdati@um.ac.ir (J. Vahdati Khaki).

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Received 31 July 2022; Received in revised form 24 August 2022; Accepted 28 August 2022 Available online 30 August 2022 2352-4928/© 2022 Elsevier Ltd. All rights reserved. [12]. Now the question is if the combination of MD simulation and the result of  $S_{conf}$  may demonstrate an exact relation between glass properties and its entropy to answer the question; it's explored by simulating four silicate-based glass, including various (1-x) SiO<sub>2</sub>-xMO, where M is alkaline earth ions (Mg<sup>2+</sup>, Ca<sup>2+</sup>, Sr<sup>2+</sup>, and Ba<sup>2+</sup> with x = 0.25, 0.30, and 35). The estimated T<sub>g</sub> values of the simulated glasses were compared with the experimental ones considering the entropy concept. The effect of additives on the concentration of NBOs, atomic distribution, Si-O, and M-O distances was investigated. The correlation between the properties and structure of glass was then discussed, considering the entropy concept. The results of this study could open new windows to researchers in the fields of glass.

## 2. Computational details

The designed silicate-based glass compositions containing various amounts (25, 30, and 35 mol%) of alkaline earth oxides (MgO, CaO, SrO, and BaO), which were studied, are listed in Table 1. The density of each glass was calculated using the Priven technique [21–23]. According to the calculated density values, about 2000 atoms were randomly entered the cubic cell with an edge of 28.8–31.98 Å according to the target system using Materials studio software (Accelrys Software Inc., San Diego, CA, USA).

The combination of long-range coulomb potential and short-range interaction in the Buckingham form was used (Eq. 1) [21].

$$U_{ij}(\mathbf{r}) = \frac{q_i q_i}{4\pi\epsilon_0 r} + A_{ij} \exp\left(\frac{\mathbf{r}}{\rho_{ij}}\right) - \frac{C_{ij}}{r^6}$$
(1)

where  $q_i$  and  $q_j$  are the atom charges and  $A_{ij}$ ,  $\rho_{ij}$ , and  $C_{ij}$  are the Buckingham potential parameters.  $\varepsilon_0$  and r are the dielectric constant of vacuum and interatomic distance, respectively. The Buckingham interatomic potential parameters are presented in Table 2 [24–26]. In addition, the intra-tetrahedral O-Si-O angles were controlled using three-body screened harmonic potential (Eq. 2):

$$U(\theta_{ijk}) = \frac{1}{2} K_{ijk}(\theta_{ijk}, \theta_{ijk,0})^2 exp(-[\frac{r_{ij}}{\rho} + \frac{r_{jk}}{\rho}])$$
(2)

where Kijk is the force constant and  $\theta ijk$ ,  $\theta$  is the reference angle of the i-j-k triplet. The mentioned parameters of three-body potential are presented in Table 2.

The MD simulations were performed using DL\_POLY\_2.18 [27]. According to the Verlet algorithm, the cutoff distance equal to 12 Å with a time step of 1 fs was used for short-range interactions. The Ewald summation method with a relative precision of  $10^{-6}$  was used to calculate the long-range Columbic interactions. To control the temperature and pressure of simulations, the Nosé -Hoover thermostat and the Andersen barostat were used, respectively. For glass simulations,

Table 1

The glass composition details for simulating the different silicate-based systems containing 25, 30, and 35 mol% additives (MgO, CaO, SrO, and BaO).

Composition	Number of species in the simulation cells		Total number of species	Cell size (A)	
	$SiO_2$	Additive			
75SiO <sub>2</sub> -25MgO	464	230	1852	28.80	
70SiO <sub>2</sub> -30MgO	442	283	1892	29.01	
65SiO <sub>2</sub> -35MgO	411	330	1893	29.12	
75SiO <sub>2</sub> -25CaO	512	183	1902	29.77	
70SiO <sub>2</sub> -30CaO	484	222	1896	29.89	
65SiO <sub>2</sub> -35CaO	452	261	1878	29.96	
75SiO <sub>2</sub> -25SrO	565	109	1913	30.76	
70SiO <sub>2</sub> -30SrO	548	136	1916	31.16	
65SiO <sub>2</sub> -35SrO	518	162	1878	31.35	
75SiO <sub>2</sub> -25BaO	591	77	1927	31.22	
70SiO <sub>2</sub> -30BaO	554	93	1848	31.28	
65SiO <sub>2</sub> -35BaO	550	116	1882	31.98	

The interatomic potential parameters [24–26].
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	1				
	Buckingham A <sub>ij</sub> ex	$p\Bigl(\frac{r}{\rho_{ij}}\Bigr) - \frac{C_{ij}}{r^6}$			
Pair atoms	A <sub>ij</sub> (eV)	ρ <sub>ij</sub> (Å)	C <sub>ij</sub> (eV. Å <sup>6</sup> )		
Si-O	13702.905	0.193817	54.681		
0-0	2029.2204	0.343645	192.58		
Mg-O	1279.69	0.2997	0.00		
Ca-O	7747.1834	0.252623	93.109		
Sr-O	14566.637	0.245015	81.773		
Ba-O	1214.4	0.3522	0.00		
	$\label{eq:three-body potential U} \begin{split} \text{Three-body potential U} (\theta_{ijk}) \ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ r_{\text{Tr}} \ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk, \ 0})^2 exp(\ - \left[\frac{r_{ij}}{\rho} + \right] \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk}) (\theta_{ijk} - \theta_{ijk}) \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk}) (\theta_{ijk} - \theta_{ijk}) (\theta_{ijk} - \theta_{ijk}) (\theta_{ijk} - \theta_{ijk}) \\ &= \frac{1}{2} K_{ijk} (\theta_{ijk} - \theta_{ijk}) (\theta_{ijk} - \theta_{ijk$				
	<u>- jk</u> ])				
	$K_{ijk}$ (eV. Rad <sup>-2</sup> )	θ <sub>ijk.0</sub> (deg)	ρ (Å)		
O-Si-O	100	109.47	1.0		

initially, the system's energy reached the minimum at 0 K and then the configurations were relaxed at 300 K for 60 ps in the NPT ensemble. The systems were melted at 4000 K for 100 ps in the NPT ensemble to ensure a suitable melting of samples. Then the melted systems were cooled at a rate of 10 K/ps to 300 K. The systems were furthermore equilibrated for 5 ns using the NPT ensemble. Finally, the production runs were performed for 5 ns, in the microcanonical ensemble (NVE) and the structural analysis was carried out on all configurations extracted from the NVE trajectory. The schematic representation of the used protocol is shown in Fig. 1. The ball-and-stick model of the extracted glasses from the molecular dynamics runs shown in Fig. 2.

The  $T_g$  of the systems was estimated using Volume-temperature curves as suggested in [16]. Besides, the experimental  $T_g$  ranges of the glass were adapted from the Sci-glass database.

Eq. 3 was used to investigate the Si-O and M-O (M = MgO, CaO, SrO, and BaO) pair distribution correlation from radial distribution function (RDF) analysis [28–30].

$$g(r) = \frac{\rho_{\text{local}}}{\rho} = \frac{dN(r)/Vdr}{\rho}$$
(3)

where g (r) is RDF or the ratio of the local density ( $\rho_{local}$ ) to the bulk density ( $\rho$ ), N is the number of atoms, and V is the total system volumes.



Fig. 1. The schematic of the used MD protocol to obtain the glass structures.



Fig. 2. The ball-and-stick model of the glasses containing 25 mol% of additives.

## 3. Results and discussion

## 3.1. The relation between $T_g$ and entropy of the simulated glasses

In the previously published study, an indirect linear relation between  $T_g$  and entropy was observed (Fig. 3) [12]. It was found that the slope of

 $T_g$  versus  $S_{conf}$  changes from one glass to another and its slopes directly correlate with alkali cation sizes as  $SiO_2\text{-}BaO < SiO_2\text{-}SrO < SiO_2\text{-}MgO < SiO_2\text{-}CaO$ . According to the list trend, it can be stated that when the entropy increases,  $T_g$  decreases (a linear relation between  $T_g$  and  $S_{conf}$  with a negative slope).

Fig. 4 shows the volume-temperature curves of the simulated glass systems.  $T_g$  of the systems was estimated considering the slope changes of the curves, and the obtained data are represented in Table 3. Besides, the adapted experimental ranges of  $T_g$  from the SciGlass database and the calculated entropy values of these systems are represented in Table 3.

According to the data, the calculated  $T_g$  of the systems changes in the range of 2956–3486 K. Increasing the mol fraction of MgO, CaO, SrO, and BaO additives from 0.25 to 0.35 decreases the  $T_g$  from 3285, 3347, 3126, and 3486–3087, 3126, 2956, and 3389 K, respectively. The decreasing trend of  $T_g$  with increasing mol fraction of alkaline earth oxides is also observed in the adapted experimental data of the SciGlass database. A significant difference between the calculated and experimental values of  $T_g$  is related to obtaining a semi-infinite solid from the simulations, as reported in [31]. As can be seen in Table 3, the calculated entropy of systems containing 25, 30, and 35 mol% additives are 0.56 R, 0.61 R, and 0.64 R, respectively. According to the data, Fig. 3, and reference [12], it can be concluded that the linear dependence of  $T_g$  with S<sub>conf</sub> is characteristic of silicate-based glasses containing alkaline earth oxides.

#### 3.2. The relation between entropy and glass structure

Adding additives to the glass structure could change the bond length and leads to lattice distortion in the glass due to the anti-polarization effect of ions [32,33]. As a result, a change in structural disordering and entropy of glass is expected. The RDF of Si-O and M-O are shown in Figs. 5 and 6, respectively. According to these figures, a short-range order glass with a modified random network (MRN) model was obtained that is in accordance with Greaves et al. study [34].

The obtained data from the RDF graphs containing positions, bond length, full width at half maximum (FWHM), and the intensity of the



Fig. 3. The linear relation with a negative slope between entropy and  $T_g$  in the silicate-based glasses containing alkaline earth oxides. Reproduced with permission from [12].



Fig. 4. Volume-temperature curves of the studied systems.

first RDF peaks was calculated and presented in Table 4. According to this table, the distance between Si and O ( $r_{Si-O}$ ) increases with increasing the amounts of additives. In the silicate system containing 25–35 mol% of MgO, CaO, SrO, and BaO, the bond length of Si-O changes in the range of 1.580–1.590, 1.591–1.596, 1.608–1.616, and 1.615–1.621 Å, respectively. Obviously, the bond length values of the elements with greater ionic radius are longer ( $Ba^{2+} > Sr^{2+} > Ca^{2+} > Mg^{2+}$ ). Increasing the bond length leads to decreasing the bond strength. In other words, adding the alkaline oxides to the glass decreases the bond strength and increases the structural disordering or entropy of systems.

In contrast,  $r_{M-O}$  of the systems mentioned above decreases with increasing the amounts of additive up to 35 mol% from 2.490, 2.491,

2.505, and 2.519–2.482, 2.484, 2.496, and 2.508 Å. Accordingly, the bond strength of glass modifiers increases with increasing their concentrations. As can be found, changing the bond length of atoms of glass former (here Si-O) is overcome other atoms' roles (here M-O) in the glass disordering. In the concept of entropy,  $S_{conf}$  of a system with one species is zero. The  $S_{conf}$  reaches to highest (0.69 R) in a binary system that has similar amounts (50–50) of species. Accordingly, the bond length/ strength of the glass former (here SiO<sub>2</sub>) primarily could affect entropy.

As shown in the RDF graphs, the intensity of Si-O correlation decreases with increasing the amounts of MgO, CaO, SrO, and BaO, in the range of 25–35 mol% from 17.66, 16.98, 15.86, 14.62–15.11, 14.76, 14.13, and 14.01, respectively. The FWHM of the mentioned samples

### Table 3

The  $T_g$  (calculated and experimental) and the  $S_{conf}$  (calculated) of the systems.

Glass Composition	T <sub>g (Calculated)</sub> (K)	T <sub>g (Exp)</sub> (K)	S <sub>conf</sub> (Calculated)
75SiO <sub>2</sub> -25MgO	3285	1063-1103	0.56 R
70SiO <sub>2</sub> -30MgO	3156	983-1015	0.61 R
65SiO <sub>2</sub> -35MgO	3087	917–933	0.64 R
75SiO <sub>2</sub> -25CaO	3347	1043-1093	0.56 R
70SiO <sub>2</sub> -30CaO	3242	1023-1045	0.61 R
65SiO <sub>2</sub> -35CaO	3126	983-1008	0.64 R
75SiO <sub>2</sub> -25SrO	3126	1046-1075	0.56 R
70SiO <sub>2</sub> -30SrO	3119	1005-1035	0.61 R
65SiO <sub>2</sub> -35SrO	2956	988-1003	0.64 R
75SiO <sub>2</sub> -25BaO	3486	1043-1088	0.56 R
70SiO <sub>2</sub> -30BaO	3422	1003-1038	0.61 R
65SiO <sub>2</sub> -35BaO	3389	1000–1057	0.64 R

increases. This observation exhibits that increasing the amounts of glass modifiers (here, alkaline earth oxides) could increase the mobility of the atoms, and increase the glass disordering, i.e., increase the  $S_{conf}$ . As can be seen, the RDF intensity of M-O increases with increasing the concentration of additives. While its FWHM experiences a reduction. Accordingly, the mobility of metal oxides decreases with increasing the concentration of additives. As discussed, the increasing mobility of Si-O glass former overcomes the decreasing mobility of MO in this situation.

### 3.3. The relation of entropy and non-bridging oxygen

All oxygens are bridging in the pristine glass, SiO<sub>2</sub>. As mentioned in the previous sections, adding species to glass leads to changing the bond

length as well as strength. In fact, adding the components to the glass structure leads to the formation of non-bridging oxygens (NBOs) [35, 36]. Previously, Du et al. showed that the concentration of NBOs in silicate-glass increases by adding alkali oxides (Li<sub>2</sub>O, Na<sub>2</sub>O, K<sub>2</sub>O) [35]. They found that the trend of NBOs concentration in different systems is  $SiO_2-Li_2O < SiO_2-Na_2O < SiO_2-K_2O$  (Fig. 7A). Also, similar trends were found in the slope of entropy-T<sub>g</sub> curves of these samples, as reported by Mollaei et al. [12]. In the current study, the concentration of NBOs of the silicate glass containing alkaline earth oxides was calculated and presented in Table 4. According to the data, the concentration of NBOs in the silicate glass containing 25–35 mol% of MgO, CaO, SrO, and BaO are in the range of 35.9–56.3, 36.8–57.5, 26.1–43.9, and 18.2–29.7. Accordingly, the trend for increasing the concentration of NBOs is  $SiO_2-BaO < SiO_2-SrO < SiO_2-MgO < SiO_2-CaO$  (Fig. 7B). Similar trends were also observed for the entropy-T<sub>g</sub> slope of these samples (Fig. 3).

When glass modifiers (e.g., alkali and alkaline earth oxides) are introduced to the glass structure, the glass network is partly destroyed, and NBOs are formed [36]. Also, the ions of modifiers (e.g.,  $Mg^{2+}$  and  $Ca^{2+}$ ) trap around the NBOs [37]. The mixing of glass modifiers with glass formers also results in the enhancement of ionic conductivity and transport properties due to the mixed anion effect [37]. All changes in the glass network at the effect of modifier additives are mainly related to changing the binding energy of glass formers with cations [37]. In summary, adding a glass modifier to the glass network leads to structural disordering or increasing the entropy of glass.



Fig. 5. RDF results of Si-O in different systems.



Fig. 6. RDF results of M-O (M = Mg, Ca, Sr, and Ba) in different systems.

Table 4 The obtained peak position, peak intensity, FWHM, and bond length of different systems.

Glass Composition	r <sub>Si-O</sub> (Å)	r <sub>M-O</sub> (Å)	Peak intensity		FWHM of peak		NBOs (%)
			Si-O	M-O	Si-O	M-O	
75SiO <sub>2</sub> -25MgO	1.580	2.490	17.66	3.03	0.25468	0.31602	35.9
70SiO <sub>2</sub> -30MgO	1.586	2.487	16.29	3.08	0.25574	0.31012	49.6
65SiO <sub>2</sub> -35MgO	1.590	2.482	15.11	3.19	0.26748	0.30589	56.3
75SiO <sub>2</sub> -25CaO	1.591	2.491	16.98	2.39	0.25987	0.33356	36.8
70SiO <sub>2</sub> -30CaO	1.593	2.488	15.24	2.43	0.26123	0.34178	49.3
65SiO <sub>2</sub> -35CaO	1.596	2.484	14.76	2.89	0.26856	0.34899	57.5
75SiO <sub>2</sub> -25SrO	1.608	2.505	15.86	2.55	0.26085	0.37807	26.1
70SiO <sub>2</sub> -30SrO	1.611	2.500	15.05	2.44	0.26743	0.37974	33.1
65SiO <sub>2</sub> -35SrO	1.616	2.496	14.13	2.39	0.27198	0.36216	43.9
75SiO <sub>2</sub> -25BaO	1.615	2.519	14.62	2.16	0.27005	0.39757	18.2
70SiO <sub>2</sub> -30BaO	1.617	2.517	14.23	2.31	0.27462	0.39612	22.6
65SiO <sub>2</sub> -35BaO	1.621	2.508	14.01	2.36	0.27891	0.38746	29.7

## 4. Conclusions

The current study successfully approved configurational entropy relation with glass structure and alkaline earth oxides composition. Different silicate-based systems containing various amounts (25, 30, and 35 mol%) of alkaline earth oxides (MO = MgO, CaO, SrO, and BaO) were initially simulated. Then, the T<sub>g</sub>, bond length, and concentration of NBOs were calculated using MD simulation. The results demonstrate that adding and increasing the concentration of alkaline earth oxides could decrease T<sub>g</sub>, increase bond length, and improve the mobility of Si-O. In addition, the concentration of NBOs increases with adding alkaline

earth oxides as well as increasing the concentration of additives. All of the mentioned parameters lead to the disordering of the glass structure and show an enhancement in the configurational entropy of systems. In conclusion, the explored relations between the configurational entropy, composition, structure, and properties of glass structures are approved in the current study. The authors hope that the proposed input, configurational entropy, is used in designing new glass materials with improved properties such as more acceptable biological performance.



Fig. 7. The variation of NBOs in percent as a function of alkali (A) and alkaline earth oxides (B). Fig. 7A Reproduced with permission from [35].

### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data Availability

Data will be made available on request.

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### Authors statement

All authors contributed to conception and design of study.

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