

## COMMUNICATION

The used potential fields and forcefield parameters utilized in this study are outlined as follows:

The potential parameters used in AAMD simulations are listed in Table S1. The atomic dynamics within the molten glass matrix are primarily dictated by the two-body BMH potential<sup>S1, S2</sup>. The pair potential, denoted as  $U_{ij}(r_{ij})$ , describes the interaction between atoms  $i$  and  $j$  as a function of their separation distance  $r_{ij}$ . The expression of  $U_{ij}(r_{ij})$  is described as:

$$U_{ij}(r_{ij}) = \frac{e^2 q_i q_j}{4\pi\epsilon_0 r_{ij}} + A_{ij} \exp\left(\frac{-r_{ij}}{B_{ij}}\right) - \frac{C_{ij}}{r_{ij}^6} + \frac{D_{ij}}{r_{ij}^{12}} \quad (1)$$

where  $q$  represents the effective charge associated with each atom,  $\epsilon_0$  denotes the dielectric constant, and  $A_{ij}$ ,  $B_{ij}$ ,  $C_{ij}$ , and  $D_{ij}$  represent force constants governing interactions between atom pairs.

In the simulation, the effective charge assigned to the oxygen atom is -0.945, while the charges for other atomic species are derived utilizing a prescribed charge allocation methodology, as delineated in previous works<sup>S3</sup>. The potential field comprises four main components: the long-range Coulombic interaction, the van der Waals term, short-range Born repulsion interactions, and an unphysical attraction occurring at distances less than 1 Å. These components collectively govern the interactions within the system. The interactions between carbon and oxygen atoms (C and O\*, respectively) encompass both electrostatic and short-range interactions. Here O\* denotes the oxygen atom within the CO<sub>2</sub>, distinguishing it from the oxygen atom (O) found in the melt. The following expression defines the potential energy  $U(1,2)$  governing the interaction between 1 and 2 of CO<sub>2</sub> molecules:

$$U(1,2) = \sum_{i \in 1} \sum_{j \in 2} \left[ 4\epsilon_{ij} \left[ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^6 \right] + \frac{e^2 q_i q_j}{4\pi\epsilon_0 r_{ij}} \right] \quad (2)$$

where  $q_C$  and  $q_{O^*}$  represent the effective charges on C and O\* atoms, respectively, with values of 0.5888 and -0.2944. Additionally,  $\sigma_{ij}$  and  $r_{ij}$  denote the Lennard-Jones potential parameters governing interactions between the C-C, C-O\*, and O\*-O\* pairs.

The intramolecular potential, employing a harmonic form to accommodate molecular flexibility, is applied to faithfully reproduce the bending vibration of CO<sub>2</sub>. This potential is described in detail in reference<sup>S4</sup>:

$$U_\theta = 1/2 k_\theta (\theta - \pi)^2 \quad (3)$$

where  $k_\theta$  represents the fixed force parameter and  $\theta$  signifies the bending angle.

The potential energy is used to describe the interactions between a CO<sub>2</sub> molecule and the cation ( $X_j$ ) within the melt. The definition of potential energy is described by the following expression:

$$U_{CO_2-X_j} = \sum_{i \in [CO_2]} \left[ \frac{e^2 q_i q_j}{4\pi\epsilon_0 r_{ij}} + A_{ij} \exp\left(\frac{-r_{ij}}{B_{ij}}\right) - \frac{C_{ij}}{r_{ij}^6} + \frac{D_{ij}}{r_{ij}^{12}} \right] \quad (4)$$

where  $i$  represents the atoms of CO<sub>2</sub> molecule and  $j$  covers all cations in melt.

The interactions between the C and O atoms are governed by the Morse potential, as detailed in reference<sup>S4</sup>:

$$U_{C-O} = D_e \left[ \left( 1 - e^{-(r-l)/\lambda} \right)^2 - 1 \right] \quad (5)$$

where  $D_e$  represents the dissociation energy of carbon-oxygen (C-O) bond;  $l$  denotes the equilibrium distance of carbon-oxygen (C-O) bond;  $\lambda$  denotes the effective width of the potential.

All mentioned force field parameters have been valid by prior publications and are readily available in our preceding article.

The computation of Pair Distribution Function (PDF) is integral to characterizes the spherically averaged local organization surrounding a specific atom. The computation equation is defined as<sup>S5</sup>:

$$g_{ij}(r) = \frac{V}{N_i N_j} \sum_j \frac{\left\langle n_{ij} \left( r - \frac{\Delta r}{2}, r + \frac{\Delta r}{2} \right) \right\rangle}{4\pi r^2 \Delta r} \quad (6)$$

where  $N_{i(j)}$  denotes the total number of atoms of type  $i$  ( $j$ ), and for atoms of the same species  $N_{i(j)} = N_i - 1$ ;  $V$  represents the volume of the simulation box;  $n_{ij}\left(r - \frac{\Delta r}{2}, r + \frac{\Delta r}{2}\right)$  is the average number of atoms  $j$  surrounding atom  $i$  within the distance  $\left(r - \frac{\Delta r}{2}, r + \frac{\Delta r}{2}\right)$ .

In the analysis of each atomic pair, the initial peak evident in their respective PDF profiles signifies the distribution of distances within the first-neighbor shell. Particularly notable in the T-O pair context, the subsequent minimum following this initial peak, identified as the first valley of the PDF curve, serves as a reliable marker for the upper limit of this distribution. Specifically, in the case of the T-O pair, a subsequent minimum following this initial peak, termed the first valley of the PDF curve, serves as a robust indicator of the upper limit of this distribution. The identification of this upper limit, referred to as the cut-off distance, holds critical significance in the determination of both the T-O coordination number and the proportion of  $\text{TO}_n$  polyhedral structures. The average coordination number (CN) can be obtained by integrating the associated PDF curve up to the cut-off value, as outlined below<sup>55</sup>:

$$N_{ij}(r) = \frac{4\pi N_j}{V} \int_0^r r^2 g_{ij}(r) dr \quad (7)$$

The analysis of mean square displacement (MSD) and self-diffusion coefficient (D) offers valuable insights into the transport behavior in the melt. The self-diffusion coefficient is determined from the MSD using the Einstein relation as<sup>56</sup>:

$$D = \frac{1}{6L} \text{MSD} = \frac{1}{6LNM} \sum_1^N \sum_k^M \langle |r_i(t_k + L) - r_i(t_k)|^2 \rangle \quad (8)$$

where  $\langle |r_i(t_k + L) - r_i(t_k)|^2 \rangle$  represents an average at time  $L$  to be taken with different time origins ( $t_k$ ) of  $k^{\text{th}}$  time series data;  $M$  denotes the number of time series data.

Table S1 Potential parameters used in AAMD simulations.

I	Silicate potential			
	$A_{ij}$ (eV)	$B_{ij}$ (Å)	$C_{ij}$ (eV/Å <sup>6</sup> )	$D_{ij}$ (eV/Å <sup>12</sup> )
Ca <sup>0.945</sup> -O <sup>-0.945</sup>	155356.043	0.178	42.2587	0
Si <sup>1.89</sup> -O <sup>-0.945</sup>	50186.0509	0.161	46.2967	0
Al <sup>1.4175</sup> -O <sup>-0.945</sup>	28482.1454	0.172	34.577	0
Na <sup>0.4725</sup> -O <sup>-0.945</sup>	145402.3125	0.178	18.8075875	0
O <sup>-0.945</sup> -O <sup>-0.945</sup>	6479.68212	0.276	85.0902	0
II	CO <sub>2</sub> intramolecular potential: $t_{c-o^*} = 1.104 \text{ Å}$ ; $k_{\theta} = 4.6096 \text{ eV/rad}$			
III	CO <sub>2</sub> -CO <sub>2</sub> intermolecular potential			
	$\epsilon$ (eV)	$\sigma$ (Å)		
C <sup>0.5888</sup> -C <sup>0.5888</sup>	0.002490	2.792		
C <sup>0.5888</sup> -O <sup>*-0.2944</sup>	0.004214	2.896		
O <sup>*-0.2944</sup> -O <sup>*-0.2944</sup>	0.007135	3.000		
IV	More potential between C of CO <sub>2</sub> and O of silicate			
	$D_e$ (eV)	$l$ (Å)	$\lambda$ (Å)	
C-O	5.0249	1.162	0.2	
V	CO <sub>2</sub> -silicate potential			
	$A_{ij}$ (eV)	$B_{ij}$ (Å)	$C_{ij}$ (eV/Å <sup>6</sup> )	$D_{ij}$ (eV/Å <sup>12</sup> )
O <sup>-0.945</sup> -O <sup>*-0.2944</sup>	3239.84106	0.276	42.5451	0
Ca <sup>0.945</sup> -O <sup>*-0.2944</sup>	155356.043	0.178	42.2587	0
Si <sup>1.89</sup> -O <sup>*-0.2944</sup>	50186.0509	0.161	46.2967	0
Al <sup>1.4175</sup> -O <sup>*-0.2944</sup>	28482.1454	0.172	34.577	0
Na <sup>0.4725</sup> -O <sup>*-0.2944</sup>	145402.3125	0.178	18.8075875	0

## References

- S1 K. Li, H. Li, C. Jiang, J. Zhang, Z. Liu and S. Ren, *Mol. Simul.*, 2020, 46, 289–299.  
S2 Y. Wang, T. Sakamaki, L. B. Skinner, Z. Jing, T. Yu, Y. Kono, C. Park, G. Shen, M. L. Rivers and S. R. Sutton, *Nat. Commun.*, 2014, 5, 1–10.  
S3 K. Zheng, Z. Zhang, F. Yang and S. Sridhar, *ISIJ Int.*, 2012, 52, 342–349.  
S4 B. Guillot and N. Sator, *Geochim. Cosmochim. Acta*, 2011, 75, 1829–1857.

- S5 Z. Piao, L. Zhu, X. Wang, Z. Liu, B. Wang, P. Xiao and Z. Yuan, *Metall. Mater. Trans. B Process Metall. Mater. Process. Sci.*, 2020, 51, 2119–2130.
- S6 C. Kim, O. Borodin and G. E. Karniadakis, *J. Comput. Phys.*, 2015, 302, 485–508.