Bobo Bai

Multicomponent diffusion in natural silicate melts: 
Toward a universal eigenvector matrix

submitted in partial fulfillment of the requirements for the degree of 
Master of Science in Earth and Environmental Sciences 
Department of Earth and Environmental Sciences 
The University of Michigan

Accepted by:

________________________  __________________________
Name                  Date

________________________  __________________________
Name                  Date

________________________  __________________________
Name                  Date

I hereby grant the University of Michigan, its heirs and assigns, the non-exclusive right to reproduce and distribute single copies of my thesis, in whole or in part, in any format. I represent and warrant to the University of Michigan that the thesis is an original work, does not infringe or violate any rights of others, and that I make these grants as the sole owner of the rights to my thesis. I understand that I will not receive royalties for any reproduction of this thesis.

☑️  Permission granted.

☐  Permission granted to copy after: _________________

☐  Permission declined.

________________________
Author Signature
ABSTRACT

Diffusion is due to thermally activated random motion of particles on the atomic scale and it is one of the two ways of mass transport (the other one is convection). In a multicomponent system, the different components interact with each other. Therefore, the concentration gradient of one component can affect not only its own motion, but also the motion of other components, leading to cross effects. As a result, diffusion in an $N$-component system is described by an $N-1$ square matrix called diffusion matrix ($[D]$). Natural silicate melt usually contains 6-10 major components (defined to be more than 1 wt% oxide), thus diffusion in natural silicate melt is always multicomponent diffusion. Previous studies show that eigenvectors of $[D]$ are roughly independent of temperature, whereas eigenvalues of $[D]$ follow an Arrhenius relation with temperature. In this study, whether the diffusion eigenvectors depend on melt composition was evaluated. MATLAB codes were written to obtain a universal eigenvector matrix by simultaneously fitting concentration profiles from 27 diffusion couple experiments in basaltic melts (Guo and Zhang, 2018, 2020). Using the MATLAB codes, the diffusion profiles of 27 diffusion couple experiments were fitted twice—using mass fraction and mole fractions as concentration, respectively—and two different eigenvector matrices $[P_w]$ and $[P_X]$ were obtained. The eigenvector matrix $[P_w]$ is significantly different from that of Guo and Zhang (2020), and our fitting more rigorously proved that diffusion eigenvectors are independent of temperature. Using both eigenvector matrices, the oxide concentrations were transformed to eigen-component “concentrations”, which were plotted as a function of distance. Then, literature diffusion profiles in diffusion couple and mineral dissolution experiments of rhyolitic to andesitic to haplo-basaltic melts were examined in our new eigen-component plots to evaluate whether the profiles are monotonic. If the eigenvector matrix from the 27 experiments is
composition-invariant, then all eigen-component “concentration” plots should be monotonic. Most eigen-component “concentration” plots are monotonic (no obvious uphill diffusion). Only about 3% of the >1000 eigen-component “concentrations” plots display obvious non-monotonic profiles, showing that we are getting close to a universal eigenvector matrix.
ACKNOWLEDGEMENT

I would like to express my sincere gratitude to my advisor, Youxue Zhang for his patience, guidance, advice, and encouragement. Without his help, I cannot complete my thesis. He has a pure passion for science and rigorous attitude towards scientific research, for which I greatly admire. I would also like to thank the other faculty reader, Jie (Jackie) Li, for her valuable suggestions and comments that greatly improve my thesis.

I would like to thank Zhengjiu Xu, for her systematic training for different kinds of experiments and measurements. Her meticulous attitude towards experiments has left a deep impression on me. I would also like to thank Owen Neil for his support on Electron Microprobe measurements.

Finally, I would like to thank all my friends and families. Especially I would like to thank my parents, Mingyang Bai and Xinhua Ma for their endless care, encouragement, and continuous support.
# Table of Contents

ABSTRACT .................................................................................................................................................................. ii

ACKNOWLEDGEMENT .......................................................................................................................................... iv

LIST OF FIGURES ..................................................................................................................................................... vi

LIST OF TABLES .................................................................................................................................................... xvii

LIST OF APPENDICES ........................................................................................................................................ xviii

I. Introduction ............................................................................................................................................................... 1

II. Theoretical background .......................................................................................................................................... 4

2.1 Diffusivity matrices, eigenvectors, and eigenvalues ............................................................................................ 4

2.2 Eigen-components and eigen-space ..................................................................................................................... 5

2.3 Analytical solutions to multicomponent diffusion ............................................................................................... 6

2.4 Diffusivity matrices when other units of concentration are used ................................................................. 7

III. Methods ................................................................................................................................................................. 10

3.1 Data collection and processing ........................................................................................................................... 10

3.2 Numerical method .............................................................................................................................................. 11

IV. Results ................................................................................................................................................................... 16

4.1 Fitting diffusion profiles with mass fraction as concentration ........................................................................... 16

4.2 Fitting diffusion profiles with mole fraction as concentration ........................................................................... 21

V. Discussion ............................................................................................................................................................... 26

5.1 Comparison to eigenvector matrices from previous studies .............................................................................. 26

5.2 Comparison of \([P_w]\) and \([P_x]\) ......................................................................................................................... 26

5.3 Verification of the universality of eigenvector matrix ....................................................................................... 28

5.4 Diffusion mechanism in natural silicate melts ................................................................................................... 30

VI. Summary and Implications ................................................................................................................................. 38

References .................................................................................................................................................................... 41

Appendices .................................................................................................................................................................. 45
### LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.1. Temperature dependence of eigenvalues listed in Table 4.1</td>
<td>17</td>
</tr>
<tr>
<td>4.2. Diffusion profiles of oxide components in wt% of BS7&amp;8C</td>
<td>18</td>
</tr>
<tr>
<td>4.3. Diffusion profiles of oxide components in wt% of BS19&amp;20C</td>
<td>19</td>
</tr>
<tr>
<td>4.4. Diffusion profiles of oxide components in wt% of BS11&amp;12A</td>
<td>20</td>
</tr>
<tr>
<td>4.5. Temperature dependence of eigenvalues listed in Table 4.3</td>
<td>22</td>
</tr>
<tr>
<td>4.6. Diffusion profiles of oxide components in mole% of BS7&amp;8C</td>
<td>23</td>
</tr>
<tr>
<td>4.7. Diffusion profiles of oxide components in mole% of BS19&amp;20C</td>
<td>24</td>
</tr>
<tr>
<td>4.8. Diffusion profiles of oxide components in mole% of BS11&amp;12A</td>
<td>25</td>
</tr>
<tr>
<td>5.1. (P_w)-converted concentration profiles of eigen-components of Zhang1989_Exp#227</td>
<td>32</td>
</tr>
<tr>
<td>5.2. (P_w)-converted concentration profiles of eigen-components of Chen2008_Exp#40</td>
<td>33</td>
</tr>
<tr>
<td>5.3. (P_w)-converted concentration profiles of eigen-components of Yu2019_Exp#106</td>
<td>34</td>
</tr>
<tr>
<td>5.4. (P_x)-converted concentration profiles of eigen-components of Zhang 1989_Exp#227</td>
<td>35</td>
</tr>
<tr>
<td>5.5. (P_x)-converted concentration profiles of eigen-components of Chen2008_Exp#40</td>
<td>36</td>
</tr>
<tr>
<td>5.6. (P_x)-converted concentration profiles of eigen-components of Yu2019_Exp#106.</td>
<td>37</td>
</tr>
<tr>
<td>6.1. Calculated diffusion profiles resulting from magma mixing between dacite and basalt</td>
<td>40</td>
</tr>
<tr>
<td>C1.1. Diffusion profiles of oxide components in wt% of BS1&amp;2C with fits</td>
<td>66</td>
</tr>
<tr>
<td>C1.2. Diffusion profiles of oxide components in wt% of BS3&amp;4C with fits</td>
<td>67</td>
</tr>
<tr>
<td>C1.3. Diffusion profiles of oxide components in wt% of BS5&amp;6C with fits</td>
<td>68</td>
</tr>
<tr>
<td>C1.4. Diffusion profiles of oxide components in wt% of BS7&amp;8C with fits</td>
<td>69</td>
</tr>
<tr>
<td>C1.5. Diffusion profiles of oxide components in wt% of BS9&amp;10C with fits</td>
<td>70</td>
</tr>
<tr>
<td>C1.6. Diffusion profiles of oxide components in wt% of BS11&amp;12C with fits</td>
<td>71</td>
</tr>
<tr>
<td>C1.7. Diffusion profiles of oxide components in wt% of BS13&amp;14C with fits</td>
<td>72</td>
</tr>
</tbody>
</table>
C1.8. Diffusion profiles of oxide components in wt% of BS17&18C with fits..........................73
C1.9. Diffusion profiles of oxide components in wt% of BS19&20C with fits.........................74
C1.10. Diffusion profiles of oxide components in wt% of BS1&2A with fits. .........................75
C1.11. Diffusion profiles of oxide components in wt% of BS3&4A with fits. .........................76
C1.12. Diffusion profiles of oxide components in wt% of BS5&6A with fits. .........................77
C1.13. Diffusion profiles of oxide components in wt% of BS7&8A with fits. .........................78
C1.14. Diffusion profiles of oxide components in wt% of BS9&10A with fits. .........................79
C1.15. Diffusion profiles of oxide components in wt% of BS11&12C with fits......................80
C1.16. Diffusion profiles of oxide components in wt% of BS13&14A with fits. .......................81
C1.17. Diffusion profiles of oxide components in wt% of BS17&18A with fits. .......................82
C1.18. Diffusion profiles of oxide components in wt% of BS19&20C with fits.......................83
C1.19. Diffusion profiles of oxide components in wt% of BS1&2B with fits.........................84
C1.20. Diffusion profiles of oxide components in wt% of BS3&4B with fits.........................85
C1.21. Diffusion profiles of oxide components in wt% of BS5&6B with fits.........................86
C1.22. Diffusion profiles of oxide components in wt% of BS7&8B with fits............................87
C1.23. Diffusion profiles of oxide components in wt% of BS9&10B with fits.........................88
C1.24. Diffusion profiles of oxide components in wt% of BS11&12B with fits......................89
C1.25. Diffusion profiles of oxide components in wt% of BS13&14B with fits......................90
C1.26. Diffusion profiles of oxide components in wt% of BS17&18B with fits......................91
C1.27. Diffusion profiles of oxide components in wt% of BS19&20B with fits......................92
C2.1. Diffusion profiles of oxide components in mole% of BS1&2C with fits.........................93
C2.2. Diffusion profiles of oxide components in mole% of BS3&4C with fits.........................94
C2.3. Diffusion profiles of oxide components in mole% of BS5&6C with fits.........................95
C2.4. Diffusion profiles of oxide components in mole% of BS7&8C with fits.........................96
D2. Concentration profiles of Zhang1989_Exp#216 ................................................................. 122
D3. Concentration profiles of Zhang1989_Exp#219 ................................................................. 123
D4. Concentration profiles of Zhang1989_Exp#220 ................................................................. 124
D5. Concentration profiles of Zhang1989_Exp#222 ................................................................. 125
D6. Concentration profiles of Zhang1989_Exp#223 ................................................................. 126
D7. Concentration profiles of Zhang1989_Exp#225 ................................................................. 127
D8. Concentration profiles of Zhang1989_Exp#226 ................................................................. 128
D9. Concentration profiles of Zhang1989_Exp#227 ................................................................. 129
D10. Concentration profiles of Zhang1989_Exp#228FO ........................................................... 130
D11. Concentration profiles of Zhang1989_Exp#228OL ........................................................... 131
D12. Concentration profiles of Zhang1989_Exp#229 ................................................................. 132
D13. Concentration profiles of Zhang1989_Exp#231DI ............................................................ 133
D15. Concentration profiles of Zhang1989_Exp#231SP ............................................................ 135
D17. Concentration profiles of Zhang1989_Exp#235OL ........................................................... 137
D18. Concentration profiles of Zhang1989_Exp#236OL ........................................................... 138
D19. Concentration profiles of Zhang1989_Exp#236RU ........................................................... 139
D20. Concentration profiles of Zhang1989_Exp#239MID OL .................................................. 140
D21. Concentration profiles of Zhang1989_Exp#242 ................................................................. 141
D22. Concentration profiles of Zhang1989_Exp#246 ................................................................. 142
D23. Concentration profiles of Zhang1989_Exp#255DI ............................................................ 143
D24. Concentration profiles of Zhang1989_Exp#255OL ........................................................... 144
D25. Concentration profiles of Zhang1989_Exp#256 ................................................................. 145
D26. Concentration profiles of Zhang1989_Exp#259AUG ........................................................ 146
D27. Concentration profiles of Zhang1989_Exp#259PL ........................................................ 147
D29. Concentration profiles of Zhang1989_Exp#260SP ........................................................ 149
D30. Concentration profiles of Zhang1989_Exp#262ENS ....................................................... 150
D31. Concentration profiles of Zhang1989_Exp#262OL ........................................................ 151
D32. Concentration profiles of Zhang1989_Exp#263OL ........................................................ 152
D33. Concentration profiles of Zhang1989_Exp#263SP ........................................................ 153
D34. Concentration profiles of Zhang1989_Exp#265 ............................................................ 154
D35. Concentration profiles of Chen2008_Exp#15 ................................................................. 155
D36. Concentration profiles of Chen2008_Exp#16 ................................................................. 156
D37. Concentration profiles of Chen2008_Exp#18 ................................................................. 157
D38. Concentration profiles of Chen2008_Exp#20 ................................................................. 158
D39. Concentration profiles of Chen2008_Exp#21 ................................................................. 159
D40. Concentration profiles of Chen2008_Exp#22 ................................................................. 160
D41. Concentration profiles of Chen2008_Exp#23 ................................................................. 161
D42. Concentration profiles of Chen2008_Exp#24 ................................................................. 162
D43. Concentration profiles of Chen2008_Exp#25 ................................................................. 163
D44. Concentration profiles of Chen2008_Exp#26 ................................................................. 164
D45. Concentration profiles of Chen2008_Exp#29 ................................................................. 165
D46. Concentration profiles of Chen2008_Exp#33 ................................................................. 166
D47. Concentration profiles of Chen2008_Exp#34 ................................................................. 167
D48. Concentration profiles of Chen2008_Exp#35 ................................................................. 168
D49. Concentration profiles of Chen2008_Exp#37 ................................................................. 169
D50. Concentration profiles of Chen2008_Exp#38 ................................................................. 170
D51. Concentration profiles of Chen2008_Exp#39 ................................................................. 171
D52. Concentration profiles of Chen2008_Exp#40 ................................................................. 172
D53. Concentration profiles of Chen2008_Exp#41 ................................................................. 173
D54. Concentration profiles of Chen2008_Exp#43 ................................................................. 174
D55. Concentration profiles of Chen2008_Exp#44 ................................................................. 175
D56. Concentration profiles of Chen2008_Exp#45 ................................................................. 176
D57. Concentration profiles of Chen2008_Exp#46 ................................................................. 177
D58. Concentration profiles of Chen2009_Exp#1 ............................................................... 178
D59. Concentration profiles of Chen2009_Exp#5 ............................................................... 179
D60. Concentration profiles of Chen2009_Exp#6 ............................................................... 180
D61. Concentration profiles of Chen2009_Exp#7 ............................................................... 181
D62. Concentration profiles of Chen2009_Exp#8 ............................................................... 182
D63. Concentration profiles of Chen2009_Exp#11 ............................................................. 183
D64. Concentration profiles of Chen2009_Exp#12 ............................................................. 184
D65. Concentration profiles of Chen2009_Exp#14 ............................................................. 185
D66. Concentration profiles of Chen2009_Exp#15 ............................................................. 186
D67. Concentration profiles of Yu2016_Exp#201 .............................................................. 187
D68. Concentration profiles of Yu2016_Exp#202 .............................................................. 188
D69. Concentration profiles of Yu2016_Exp#203 .............................................................. 189
D70. Concentration profiles of Yu2016_Exp#205 .............................................................. 190
D71. Concentration profiles of Yu2016_Exp#207 .............................................................. 191
D72. Concentration profiles of Yu2016_Exp#208 .............................................................. 192
D73. Concentration profiles of Yu2016_Exp#209 .............................................................. 193
D122. Concentration profiles of Guo&Zhang2020_BS5&6C .................................................... 242
D123. Concentration profiles of Guo&Zhang2020_BS7&8C .................................................... 243
D124. Concentration profiles of Guo&Zhang2020_BS9&10C .................................................. 244
D125. Concentration profiles of Guo&Zhang2020_BS11&12C ................................................ 245
D126. Concentration profiles of Guo&Zhang2020_BS13&14C ................................................ 246
D127. Concentration profiles of Guo&Zhang2020_BS17&18C ................................................ 247
D128. Concentration profiles of Guo&Zhang2020_BS19&20C ................................................ 248
D129. Concentration profiles of Guo&Zhang2018_BS1&2A .................................................... 249
D130. Concentration profiles of Guo&Zhang2018_BS3&4A .................................................... 250
D131. Concentration profiles of Guo&Zhang2018_BS5&6A .................................................... 251
D132. Concentration profiles of Guo&Zhang2018_BS7&8A .................................................... 252
D133. Concentration profiles of Guo&Zhang2018_BS9&10A .................................................. 253
D134. Concentration profiles of Guo&Zhang2018_BS11&12A ................................................ 254
D135. Concentration profiles of Guo&Zhang2018_BS13&14A ................................................ 255
D136. Concentration profiles of Guo&Zhang2018_BS17&18A ................................................ 256
D137. Concentration profiles of Guo&Zhang2018_BS19&20A ................................................ 257
D138. Concentration profiles of Guo&Zhang2020_BS1&2B .................................................... 258
D139. Concentration profiles of Guo&Zhang2020_BS3&4B .................................................... 259
D140. Concentration profiles of Guo&Zhang2020_BS5&6B .................................................... 260
D141. Concentration profiles of Guo&Zhang2020_BS7&8B .................................................... 261
D142. Concentration profiles of Guo&Zhang2020_BS9&10B .................................................. 262
D143. Concentration profiles of Guo&Zhang2020_BS11&12B ................................................ 263
D144. Concentration profiles of Guo&Zhang2020_BS13&14B ................................................ 264
D145. Concentration profiles of Guo&Zhang2020_BS17&18B ................................................ 265
D146. Concentration profiles of Guo&Zhang2020_BS19&20B ............................................ 266
D147. Concentration profiles of González2017_P050-H0-4 ...................................................... 267
D148. Concentration profiles of González2017_P050-H1-4 ...................................................... 268
D149. Concentration profiles of González2017_P050-H2-4 ...................................................... 269
D150. Concentration profiles of González2017_P100-H0-4 ...................................................... 270
D151. Concentration profiles of González2017_P100-H1-4 ...................................................... 271
D152. Concentration profiles of González2017_P100-H2-4 ...................................................... 272
D153. Concentration profiles of González2017_P300-H0-4 ...................................................... 273
D154. Concentration profiles of González2017_P300-H0-1 ...................................................... 274
D155. Concentration profiles of González2017_P300-H1-4 ...................................................... 275
D156. Concentration profiles of González2017_P300-H2-4 ...................................................... 276
D157. Concentration profiles of González2017_P300-H2-1 ...................................................... 277
D158. Concentration profiles of González2017_P300-H2-0 ...................................................... 278
D159. Concentration profiles of González2017_P300-H2-0 ...................................................... 279
D160. Concentration profiles of Wang2020_Pyx3 ..................................................................... 280
D161. Concentration profiles of Wang2020_Pyx10 ................................................................... 281
D162. Concentration profiles of Wang2020_Pyx11 ................................................................... 282
D163. Concentration profiles of Wang2020_Pyx26 ................................................................... 283
D164. Concentration profiles of Wang2020_Pyx21 ................................................................... 284
D165. Concentration profiles of Wang2020_Pyx27 ................................................................... 285
D166. Concentration profiles of Wang2020_Pyx17 ................................................................... 286
D167. Concentration profiles of Wang2020_Pyx20 ................................................................... 287
D168. Concentration profiles of Wang2020_26W01 .................................................................. 288
D169. Concentration profiles of Wang2020_26W02 .................................................................. 289
D170. Concentration profiles of Wang2020_JW03 ..........................................................................................290

D171. Concentration profiles of Wang2020_JW02 ..........................................................................................291
LIST OF TABLES

Table | Page
------|------
4.1. Eigenvalues and eigenvector matrix fitted by mass fraction data. | 16
4.2. Temperature dependence of eigenvalues listed in Table 4.1 and corresponding activation energies. | 17
4.3. Eigenvalues and eigenvector matrix fitted by mole fraction data. | 21
4.4. Temperature dependence of eigenvalues listed in Table 4.3 and corresponding activation energies. | 22
5.1. The constant eigenvector matrix \([P_0]\) obtained by Guo and Zhang (2020) and \([P_w]\) obtained in this study. | 26
5.2. Eigenvector matrix \([P^*_X]\) and eigenvector matrix \([P_X]\). | 28
5.3. Summary of the 35 \([P_w]\)-converted concentration profiles of eigen-components that show obvious uphill diffusion, along with the corresponding experimental conditions. | 29
5.4. Summary of the 28 \([P_X]\)-converted concentration profiles of eigen-components that show obvious uphill diffusion, along with the corresponding experimental conditions. | 30
5.5. The inverse of the eigenvector matrix \([P_X]\). | 31
# LIST OF APPENDICES

<table>
<thead>
<tr>
<th>Appendix</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>A. Derivation of an explicit expression of Jacobian matrix</td>
<td>45</td>
</tr>
<tr>
<td>B. MATLAB codes for fitting diffusion profiles</td>
<td>52</td>
</tr>
<tr>
<td>B1. Main program “BFGS_Main.m”</td>
<td>52</td>
</tr>
<tr>
<td>B2. Subroutine “con.m”</td>
<td>58</td>
</tr>
<tr>
<td>B3. Subroutine “calculate_J.m”</td>
<td>59</td>
</tr>
<tr>
<td>B4. Subroutine “calculate_J7.m”</td>
<td>62</td>
</tr>
<tr>
<td>C. Fits for 27 diffusion couple experiments from Guo and Zhang (2018, 2020)</td>
<td>65</td>
</tr>
<tr>
<td>C1. Fits of diffusion profiles using mass fraction as concentration</td>
<td>66</td>
</tr>
<tr>
<td>C2. Fits of diffusion profiles using mole fraction as concentration</td>
<td>93</td>
</tr>
<tr>
<td>D. Concentration profiles of oxide components and eigen-components</td>
<td>120</td>
</tr>
</tbody>
</table>
I. Introduction

Natural silicate melt usually contains 6-10 major components (defined to be more than 1 wt% oxide, e.g., SiO\textsubscript{2}-TiO\textsubscript{2}-Al\textsubscript{2}O\textsubscript{3}-FeO-MgO-CaO-Na\textsubscript{2}O-K\textsubscript{2}O) and diffusion in natural silicate melt is always multicomponent diffusion. In nature, many geological processes involving mass transport are essentially multicomponent diffusion, e.g., magma mixing and contamination (Sato, 1975; Watson, 1982; Koyaguchi, 1985, 1989; Oldenburg et al., 1989), and mineral growth and dissolution (Watson, 1982; Zhang et al., 1989). To quantitatively predict mass transport requires an in-depth understanding of multicomponent diffusion in natural silicate melts.

A classic treatment for diffusion in natural silicate melts is effective binary diffusion (EBD) model (e.g., Cooper, 1968), which treats diffusant \( i \) of interest as one component and all other components as one “component” \( m \) (stands for mixture). Then the flux of component of interest is assumed to be:

\[
\mathbf{J}_i = -D_{im} \nabla C_i
\]

where \( \mathbf{J}_i \) is the flux of component \( i \), \( D_{im} \) is the effective binary diffusion coefficient (EBDC) and \( \nabla C_i \) is the concentration gradient of component \( i \). Due to its simplicity, the method has been extensively used for monotonic profiles (e.g., Zhang et al., 1989; Chen and Zhang, 2008, 2009). However, the method fails when it comes to uphill diffusion, which is often encountered in natural systems and experiments. In addition, the effective binary diffusion diffusivities depend strongly on thermodynamic condition, melt composition and the direction of diffusion in composition space, which restricts the applications of EBDCs to interdiffusion under a different thermodynamic condition and in melt systems with a different composition (Cooper, 1968; Zhang et al., 1989; Liang et al., 1996; Zhang, 2010; Chen and Zhang, 2008, 2009; Guo and Zhang, 2016, 2018, 2020).
To treat non-monotonic diffusion profiles, Zhang (1993) proposed a modified effective binary diffusion model, which assumes the diffusive flux of a component is proportional to its activity gradient (instead of concentration gradient).

\[
J_i = -\frac{D_i}{\gamma_i} \nabla a_i
\]

(2)

where \( J_i \), \( D_i \), \( a_i \) and \( \gamma_i \) are the flux, activity based EBDC (a.k.a. intrinsic EBDC), activity and activity coefficient of component \( i \), respectively. Even though the model has some values of fitting non-monotonic diffusion profiles, due the complexity of the model and lack of activity data for silicate melts, it has not been widely applied. In addition, both the EBD model and the modified EBD model neglect interactions among diffusants, making it difficult to decipher the diffusion mechanism in natural silicate melts.

To solve diffusion problems in a multicomponent system more rigorously, there is a critical need for a multicomponent treatment (Onsager, 1945; Liang, 2010; Zhang, 2010, 2022), in which the flux of the component \( i \) is a linear combination of the concentration gradients of independent components:

\[
J_i = -\sum_j D_{ij} \nabla C_j
\]

(3)

where \( J_i \) is the flux of component \( i \), \( D_{ij} \) is diffusion coefficient, which characterizes the cross effect of the concentration gradients of the component \( j \) on the flux of component \( i \), and \( \nabla C_j \) is the concentration gradient of component \( j \). The \( n \times n \) square matrix whose \((i, j)\) entry is \( D_{ij} \) is called the diffusivity matrix and denoted by \([D]\). Enormous efforts have been made to determine the diffusivity matrix at desired compositions, e.g., SiO\(_2\)-Al\(_2\)O\(_3\)-CaO (Sugawara et al., 1977; Oishi et al., 1982; Liang et al., 1996), SiO\(_2\)-Al\(_2\)O\(_3\)-MgO (Kress and Ghiorso, 1993; Richter et al., 1998), SiO\(_2\)-Al\(_2\)O\(_3\)-K\(_2\)O (Chakraborty et al., 1995a), SiO\(_2\)-B\(_2\)O\(_3\)-Na\(_2\)O (Pablo et al., 2017), SiO\(_2\)-Al\(_2\)O\(_3\)-MgO-CaO (Kress and Ghiorso, 1993), SiO\(_2\)-Al\(_2\)O\(_3\)-Na\(_2\)O-K\(_2\)O-H\(_2\)O (Mungall et al., 1998),
SiO$_2$-TiO$_2$-Al$_2$O$_3$-MgO-CaO-Na$_2$O-K$_2$O (Guo and Zhang, 2016) and SiO$_2$-TiO$_2$-Al$_2$O$_3$-FeO-MgO-CaO-Na$_2$O-K$_2$O (Guo and Zhang, 2018, 2020).

Among those studies, Guo and Zhang (2018, 2020) conducted numerous experiments to extract diffusivity matrices in an 8-component system (SiO$_2$-TiO$_2$-Al$_2$O$_3$-FeO-MgO-CaO-Na$_2$O-K$_2$O) at 1260 °C, 1350 °C and 1500 °C. Their success of using diffusivity matrix to predict mass transport during mineral dissolution shows the validity and utility of multicomponent treatment in diffusion in natural silicate melts. In addition, they found that eigenvectors of $[D]$ are roughly independent of temperature and eigenvalues of $[D]$ follows an Arrhenius relation with temperature. With these observations, we can hence calculate diffusion matrix in basaltic melt at temperatures within the studied range. Furthermore, the calculated diffusion matrix can be used to predict diffusion behavior in magma mixing and mineral dissolution. However, if the diffusion involves a large difference in melt composition (i.e., from basalt to rhyolite), it would be necessary to first assess the compositional effect on the diffusion matrix.

In this study, we focus on an 8-component melt system (SiO$_2$-TiO$_2$-Al$_2$O$_3$-FeO-MgO-CaO-Na$_2$O-K$_2$O) and evaluate whether diffusion eigenvectors depend on composition. Diffusion profiles from previous studies (e.g., Guo and Zhang 2018, 2020) were collected and fitted by assuming a constant eigenvector matrix. Then the best two eigenvector matrices—obtained by fitting diffusion profiles using mass fraction and mole fraction as concentration, respectively—were used to calculate profiles of eigen-components. Finally, the profiles of eigen-components were plotted to check whether they show uphill diffusion features.
II. Theoretical background

2.1 Diffusivity matrices, eigenvectors, and eigenvalues

In an \( N \)-component melt system with a barycentric (mass-fixed) frame of reference, without bulk motion, sink and source, there are only \( n = N - 1 \) independent components (Kirkwood et al., 1960; Miller et al., 1986). As a generalization of Fick’s first law (Onsager, 1945), the flux of the component \( i \) is a linear combination of the concentration gradients of independent components, i.e.,

\[
\mathbf{J}_i = -\sum_{j=1}^{n} D_{ij}^N \nabla C_j
\]

Here \( \mathbf{J}_i \) is mass-unit flow. \( D_{ij}^N \) is the coefficient characterizing the cross effect of the concentration gradient of the component \( j \) on the flux of component \( i \), where the superscript \( N \) means we choose the \( N^{th} \) component as the dependent component. And \( C_j \) is the concentration of component \( j \) expressed as mass per unit volume, usually with a unit of g/cm\(^3\). Accordingly, the vector form of Fick’s first law is given by:

\[
\mathbf{J} = \left[ \begin{array}{c}
J_1 \\
J_2 \\
\vdots \\
J_n \\
\end{array} \right] = \left[ \begin{array}{c}
-\sum_{j=1}^{n} D_{1j}^N \nabla C_j \\
-\sum_{j=1}^{n} D_{2j}^N \nabla C_j \\
\vdots \\
-\sum_{j=1}^{n} D_{nj}^N \nabla C_j \\
\end{array} \right] = -\left[ \begin{array}{cccc}
D_{11}^N & D_{12}^N & \cdots & D_{1n}^N \\
D_{21}^N & D_{22}^N & \cdots & D_{2n}^N \\
\vdots & \vdots & \ddots & \vdots \\
D_{n1}^N & D_{n2}^N & \cdots & D_{nn}^N \\
\end{array} \right] \times \nabla \left[ \begin{array}{c}
C_1 \\
C_2 \\
\vdots \\
C_n \\
\end{array} \right] = -[D^N] \nabla \mathbf{C} \tag{5}
\]

where \([D^N]\) is the diffusivity matrix.

It has been shown that in a thermodynamically stable region \([D^N]\) is positive-definite (e.g., Gupta and Cooper, 1971), namely, having positive eigenvalues, and hence the diagonal terms of \([D^N]\) are always positive. However, the off-diagonal terms of \([D^N]\) can be either positive or negative, and sometimes has large absolute values, which enables a component to diffuse against its concentration gradient, leading to uphill diffusion. Mathematically, \([D^N]\) can be diagonalized:

\[
[D^N] = [P][A][P^{-1}] \tag{6}
\]
where \([P]\) is an \(n \times n\) matrix whose columns \(v_i\) are eigenvectors of \([D^N]\), \([P^{-1}]\) is the inverse of \([P]\) and \([\Lambda]\) is an \(n \times n\) diagonal matrix whose diagonal terms \(\lambda_i\) are eigenvalues of \([D^N]\), such that:

\[
[D^N]v_i = \lambda_i v_i \tag{7}
\]

The eigenvectors of \([D^N]\) describe interactions among components and are commonly used to infer exchange mechanisms and the eigenvalues of \([D^N]\) characterize the exchange rates in the melt system (Varshneya and Cooper, 1972; Liang et al., 1996; Guo and Zhang, 2016, 2018, 2020).

### 2.2 Eigen-components and eigen-space

Combining Fick’s first law and mass conservation gives Fick’s second law:

\[
\frac{\partial C}{\partial t} = \nabla \cdot ([D^N]\nabla C) \tag{8}
\]

where \(t\) is time.

Diffusion problems are usually studied in one dimension; thus Eq. (8) is often simplified to the expression:

\[
\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left( [D^N] \frac{\partial C}{\partial x} \right) \tag{9}
\]

Plugging Eq. (6) into Eq. (9), leads to

\[
\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left( [P][\Lambda][P^{-1}] \frac{\partial C}{\partial x} \right) \tag{10}
\]

If eigenvector matrix \([P]\) is independent of melt composition, then Eq. (10) can be converted into the following expression:

\[
\frac{\partial [P^{-1}]C}{\partial t} = \frac{\partial}{\partial x} \left( [\Lambda] \frac{\partial [P^{-1}]C}{\partial x} \right) \tag{11}
\]

Denoting \([P^{-1}]C\) by \(Z\), namely,
\[ Z = \begin{bmatrix} Z_1 \\ Z_2 \\ \vdots \\ Z_n \end{bmatrix} = [P^{-1}] \begin{bmatrix} C_1 \\ C_2 \\ \vdots \\ C_n \end{bmatrix} = [P^{-1}]C \]  

(12)

then Eq. (11) can be converted into the following equation:

\[ \frac{\partial Z}{\partial t} = \frac{\partial}{\partial x} \left( [\Lambda] \frac{\partial Z}{\partial x} \right) \]  

(13)

Eq. (13) contains \( n \) independent equations:

\[ \frac{\partial Z_i}{\partial t} = \frac{\partial}{\partial x} \left( \lambda_i \frac{\partial Z_i}{\partial x} \right) \]  

(14)

To distinguish \( C \) from \( Z \), we name each term \( C_i \) in \( C \) an oxide component and each term \( Z_i \) in \( Z \) an eigen-component. Accordingly, the compositional space composed of \( Z_i \) is referred to as eigen-space. Note that since \( \lambda_i \) is always positive, profiles of eigen-components should always be monotonic as long as the initial concentration profile is monotonic. Since Eq. (14) is obtained based on a constant eigenvector matrix \([P]\), the monotonicity of profiles of eigen-components is a necessary condition of the universality of eigenvector matrix \([P]\).

2.3 Analytical solutions to multicomponent diffusion

For multicomponent diffusion in an infinite diffusion couple, assuming that \([D^N]\) is constant within the studied range and the interface position is at \( x = 0 \), the analytical solution can be easily obtained (Lasaga, 1979; Morgan et al., 2006; Guo and Zhang, 2016):

\[ C = \frac{C_R + C_L}{2} + [P][E][P^{-1}] \frac{C_R - C_L}{2} \]  

(15)

where \( C_L \) and \( C_R \) are initial melt concentration vectors at \( x < 0 \) and \( x > 0 \), \([P]\) is eigenvector matrix, \([E]\) is a diagonal matrix whose diagonal terms are:

\[ E_{ii} = \text{erf} \left( \frac{x}{\sqrt{4\lambda_i t}} \right) \]  

(16)

in which \( x \) is distance from the interface and \( t \) is time.
2.4 Diffusivity matrices when other units of concentration are used

The diffusivity matrix $[D^N]$ depends on many factors, e.g., thermodynamic state (e.g., Liang and Davis, 2002; Guo and Zhang, 2020), melt composition (e.g., Liang et al., 1996), the choice of the dependent component (e.g., Liang et al., 1996; Guo and Zhang, 2016, 2018, 2020), the choice of reference frame (e.g., Kirkwood et al., 1960; Miller et al., 1986), and the choice of concentration unit. Among them, the effect of choice of concentration unit hasn’t been studied. Here we derive how diffusivity matrix varies when cation mole concentration, mass fraction and cation mole fraction are used.

First, one may substitute the vector $C$ of mass concentration (kg/m$^3$) by the vector $\tilde{C}$ of cation molar concentration (mol/m$^3$), using the following expression:

$$C = \begin{bmatrix} C_1 \\ C_2 \\ \vdots \\ C_n \end{bmatrix} = \begin{bmatrix} M_1 \tilde{C}_1 \\ M_2 \tilde{C}_2 \\ \vdots \\ M_n \tilde{C}_n \end{bmatrix} = [M^N] \tilde{C}$$

(17)

where $\tilde{C}_i$ is molar concentration of the $i^{th}$ cation and $[M^N]$ is a diagonal matrix with the diagonal term $M_i$ as the molar mass of the $i^{th}$ oxide component, namely,

$$[M^N] = \begin{bmatrix} M_1 & 0 & \cdots & 0 \\ 0 & M_2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & M_n \end{bmatrix}$$

(18)

Plugging Eq. (17) into Eq. (8), leads to

$$\frac{\partial \tilde{C}}{\partial t} = \nabla \cdot \left( [M^{N^{-1}}][D^N][M^N] \nabla \tilde{C} \right)$$

(19)

where $[M^{N^{-1}}]$ is the inverse of $[M^N]$. We see from Eq. (19) that the diffusivity matrix $[\tilde{D}^N]$, when cation mole concentration is used, is related to $[D^N]$ by the following expression:

$$[\tilde{D}^N] = [M^{N^{-1}}][D^N][M^N]$$

(20)

Plugging Eq. (6) into Eq. (20), leads to:
\[
[D^N] = \left[ M^{N-1} \right] [P][\Lambda][P^{-1}][M^N] = \left( \left[ M^{N-1} \right] [P] \right) [\Lambda] \left( \left[ M^{N-1} \right] [P] \right)^{-1}
\] (21)

From Eq. (21) we see that the eigenvector matrix \([\tilde{P}]\) obtained from \(\tilde{C}\) would be:

\[
[\tilde{P}] = \left[ M^{N-1} \right] [P]
\] (22)

and the eigenvalue matrix obtained from \(\tilde{C}\) should be the same as that of \([D^N]\).

For convenience, concentration data are often expressed as mass fraction or mole fraction. In the former case, the concentration vector is often substituted by the mass fraction vector \(w\) using the equation:

\[
C = \begin{bmatrix} C_1 \\ C_2 \\ \vdots \\ C_n \end{bmatrix} = \begin{bmatrix} \rho w_1 \\ \rho w_2 \\ \vdots \\ \rho w_n \end{bmatrix} = \rho w
\] (23)

where \(\rho\) is the mass density of the melt and \(w_i\) is the mass fraction of the \(i^{th}\) oxide component.

Plugging Eq. (23) into Eq. (8) and assuming \(\rho\) doesn’t change much within the studied composition range, then we obtain the following expression:

\[
\frac{\partial w}{\partial t} = \nabla \cdot ([D^N] \nabla w)
\] (24)

Eq. (24) shows that the diffusivity matrix \([D_w^N]\) obtained from \(w\) is the same as \([D^N]\), and hence has the same eigenvector matrix \([P_w]\), namely,

\[
[D_w^N] = [D^N]
\] (25)

and

\[
[P_w] = [P]
\] (26)

Alternatively, the concentration vector \(C\) can be replaced by vector \(X\) of cation mole fraction using the expression:

\[
C = \begin{bmatrix} C_1 \\ C_2 \\ \vdots \\ C_n \end{bmatrix} = \begin{bmatrix} \tilde{\rho} M_1 X_1 \\ \tilde{\rho} M_2 X_2 \\ \vdots \\ \tilde{\rho} M_n X_n \end{bmatrix} = \tilde{\rho} [M^N] X
\] (27)
If we assume $\bar{\rho}$ is nearly constant within the studied composition range, plugging Eq. (27) into Eq. (8) leads to:

$$\frac{\partial \mathbf{X}}{\partial t} = \nabla \cdot \left( [M^{N-1}] [D^N] [M^N] \nabla \mathbf{X} \right)$$

(28)

Thus, according to Eq. (28), the diffusivity matrix $[D^N]$ obtained from $\mathbf{X}$ is:

$$[D^N] = [M^{N-1}] [D^N] [M^N]$$

(29)

By comparing Eq. (20) and Eq. (29), we see that:

$$[D^N] = \tilde{D} = \left( [M^{N-1}] [\Lambda] \left( [M^{N-1}] [P] \right)^{-1} \right)$$

(30)

Therefore, the eigenvector matrix $[P_X]$ obtained from $\mathbf{X}$ would be:

$$[P_X] = \tilde{P} = \left[ M^{N-1} \right] [P]$$

(31)

and the eigenvalue matrix obtained from $\mathbf{X}$ should be the same as those of $[D^N]$. 
III. Methods

3.1 Data collection and processing

Eigenvectors and eigenvalues in desired melt composition can be obtained by simultaneously fitting diffusion profiles of all oxides from diffusion experiments. Diffusion profiles in the 8-component melt system (SiO$_2$-TiO$_2$-Al$_2$O$_3$-FeO-MgO-CaO-Na$_2$O-K$_2$O) were collected from previous studies (Zhang et al., 1989; Chen and Zhang, 2008, 2009; Yu et al., 2016; Yu et al., 2019; Guo and Zhang, 2016, 2018, 2020; González-Garcia et al., 2017; Wang et al., 2020). The studied melt compositions vary from basaltic melt to rhyolitic melt. The oxide concentrations from those studies were all reported as mass fraction in weight percent (wt%).

Original concentration profiles were first plotted to check data quality. Then points that apparently deviate from diffusion trend or show a significant quench effect in mineral dissolution experiments were removed. After that, mass fraction concentrations $w_i$ were converted into mole fraction concentrations $X_i$ using the expression:

$$X_i = \frac{w_i}{\sum_{j=1}^{N} \frac{w_j}{M_j}} \times 100 \text{ mole}\%$$

(32)

where $w_i$ is the mass fraction of component $i$, $X_i$ is the mole fraction of component $i$, $M_i$ is the molar mass of component $i$ on a single cation basis, and $N$ is the total number of components. In our calculation, we replaced SiO$_2$ with SiO$_2^* = \text{SiO}_2 - (\text{total} - 100)$ and treated SiO$_2^*$ as the dependent component following the studies of Guo and Zhang (2016, 2018, 2020).

Only the experimental data from Guo and Zhang (2018, 2020) were incorporated in the fitting process, as those data show small concentration variations (typically less than 3 wt%) across the diffusion profiles. Those data are from diffusion couple experiments in a basaltic melt at three different temperatures, 1260 °C, 1350 °C and 1500 °C. For simplicity of experiment
naming, the three temperatures were represented by the letters C, A and B, respectively. For example, “BS1&2C” denotes a diffusion couple experiment conducted at 1260 ℃. The selected experimental data were fitted using mass fraction as concentration and mole fraction as concentration separately, generating two eigenvector matrices \([P_w]\) and \([P_x]\). After the fitting process, all collected experimental data were used to verify the universality of the extracted eigenvector matrices. Since the monotonicity of profiles of eigen-components is a necessary condition of the universality of eigenvector matrix, a natural next step is to plot profiles of eigen-components. Concentration vector \(\mathbf{w}\) (or \(\mathbf{X}\)) of oxide components was converted into concentration vector \(\mathbf{Z_w}\) (or \(\mathbf{Z_x}\)) of eigen-components using the equation:

\[
\mathbf{Z_w} = [P_w^{-1}]\mathbf{w}
\]

or

\[
\mathbf{Z_x} = [P_x^{-1}]\mathbf{X}
\]

If profiles of eigen-components are indeed monotonic, then our hypothesis is consistent with experimental data. The hypothesis can be further tested by more experimental data, but there is no straightforward proof of the hypothesis.

### 3.2 Numerical method

In this part, we do not distinguish between the fitting using mass fraction as concentration and that using mole fraction as concentration, since they are essentially the same in terms of computation. The selected diffusion couple data were fitted using one eigenvector matrix \([P]\) and three sets of eigenvalue matrices \([\Lambda^{1260}]\), \([\Lambda^{1350}]\) and \([\Lambda^{1500}]\) where the superscripts 1260, 1350 and 1500 represent temperature in ℃. To optimize eigenvectors and eigenvalues, we follow the procedure in the studies of Guo and Zhang (2016, 2018, 2020) to minimize \(\chi^2\):
\[
\chi^2 = \frac{1}{2} \sum_{k=1}^{N_e} \sum_{j=1}^{Np_k} \sum_{i=1}^{Nc} \left( \frac{w_{ijk}^{\text{meas}} - w_{ijk}^{\text{calc}}}{\sigma_{ik}} \right)^2
\]  

(35)

where \( N_c \) is the number of components (\( N_c = 8 \)), \( N_p \) is the number of points in the \( k^{th} \) experiments, \( N_e \) is number of experiments incorporated in the fitting process (\( N_e = 27 \)), \( w_{ijk}^{\text{meas}} \) is the measured concentration of oxide \( i \) at position \( j \) in experiment \( k \), \( w_{ijk}^{\text{calc}} \) is the calculated concentration using Eq. (15) and Eq. (16), \( \sigma_{ik} \) is 1-\( \sigma \) error on \( w_{ijk}^{\text{meas}} \).

Denoting \( \frac{w_{ijk}^{\text{meas}} - w_{ijk}^{\text{calc}}}{\sigma_{ik}} \) in Eq. (35) by \( r_{ijk} \) and putting all \( r_{ijk} \) in a column vector \( \mathbf{r} \) in ascending order of \((i, j, k)\) with \( k \) as primary index, \( j \) as secondary index and \( i \) as tertiary index, namely,

\[
\mathbf{r} = (r_{111}, r_{211}, ..., r_{N_c11}, r_{121}, r_{221}, ..., r_{N_c21}, ..., r_{N_cN_{p1}}, r_{112}, r_{212}, ..., r_{N_cN_{p1}N_{N_e}})^T
\]  

(36)

then Eq. (35) can be converted into the expression:

\[
\chi^2 = \frac{1}{2} \mathbf{r}^T \mathbf{r}
\]  

(37)

Our fitting parameters include 49 elements in the \( 7 \times 7 \) eigenvector matrix \([P]\) and 21 eigenvalues in 3 diagonal eigenvalue matrices at three different temperatures. One complexity is that all eigenvalues \( \lambda \) must be positive. However, if we take \( \theta = \ln \lambda \), then \( \theta \) can be any real values. Therefore, \( \theta \) is treated as the fitting parameter in place of \( \lambda \). For simplicity, we put our 70 fitting parameters into a column vector \( \mathbf{\beta} \), in which the first 49 elements are the vectorized eigenvector matrix and the remaining 21 elements are \( \theta \) values, namely,

\[
\mathbf{\beta} = (P_{11}, P_{21}, ..., P_{67}, P_{77}, \theta_1^{1260}, ..., \theta_7^{1260}, \theta_1^{1350}, ..., \theta_7^{1350}, \theta_1^{1500}, ..., \theta_7^{1500})^T
\]  

(38)

For the iterative algorithm, we used the Broyden–Fletcher–Goldfarb–Shanno method (independently proposed by Broyden, Fletcher, Goldfarb and Shanno, 1970) with the Armijo-type line search and a cautious update (Donghui and Masao, 2001b). The Broyden–Fletcher–Goldfarb–Shanno (BFGS hereafter) method is one of the most popular Quasi-Newton methods
for solving unconstrained nonlinear optimization problems. Conventional Newton method involves the calculation of Hessian matrix $[H]$—a square matrix of second-order partial derivatives of the loss function—and the inversion of $[H]$, which is very computationally expensive. In addition, to make sure the algorithm converges to a minimum, $[H]$ is required to be positive definite. When the loss function is non-convex, however, the positive definiteness of $[H]$ cannot be guaranteed. In contrast to Newton method, Quasi-Newton methods approximate $[H]$ with a positive definite matrix $[B]$, which is updated iteratively using the information calculated from previous steps. Quasi-Newton methods differ in the way they construct $[B]$, but all of them satisfy the Quasi-Newton condition. Among various Quasi-Newton methods, the BFGS method is currently regarded as the most efficient method (Donghui and Masao, 2001a) and has been widely used to solve unconstrained nonlinear optimization problems.

Generally, consider the unconstrained nonlinear optimization problem: $\min f(\beta), \beta \in \mathbb{R}^m$, where $f: \mathbb{R}^m \rightarrow \mathbb{R}$ is a continuously differentiable function (in this study $f = \chi^2$ and $m = 70$). The BFGS method begins with an initial guess for the optimal value $\beta_0$ and an initial symmetric and positive definite matrix $B_0 \in \mathbb{R}^{m \times m}$ and updates $\beta$ using the expression:

$$\beta_{k+1} = \beta_k + \gamma_k p_k, \quad k = 0, 1, 2 \ldots$$  (39)

where $\gamma_k$ is the step size at step $k$ and $p_k$ is the search direction at step $k$.

The search direction $p_k$ is given by the equation:

$$p_k = -B_k^{-1} g_k$$  (40)

where $g_k$ is the gradient of $f$ at $\beta = \beta_k$, namely,

$$g_k = \nabla f(\beta_k)$$  (41)

The step size $\gamma_k$ can be obtained by the backtracking line search which looks for the largest value $\gamma_k$ in the set $\{\rho^i | i = 0, 1 \ldots\}$ that satisfies the inequality:
\[ f(\beta_k + \gamma_k p_k) \leq f(\beta_k) + \sigma \gamma_k g_k^T p_k \]  

(42)

where \( \rho \) and \( \sigma \) are control parameters so that \( \rho, \sigma \in (0, 1) \).

The original BFGS method updates \( B_k \) using the following equation:

\[
B_{k+1} = B_k - \frac{B_k s_k s_k^T B_k}{s_k^T B_k s_k} + \frac{y_k y_k^T}{y_k^T s_k}
\]

(43)

where \( s_k = \beta_{k+1} - \beta_k \) and \( y_k = g_{k+1} - g_k \). Eq. (43) conserves the symmetry of the matrix \([B]\) and as long as \( y_k^T s_k > 0 \), \( B_{k+1} \) is also positive definite. However, the backtracking method cannot guarantee that \( y_k^T s_k > 0 \). To make sure \( B_{k+1} \) is positive definite, Donghui and Masao (2001b) proposed a cautious update that determines \( B_{k+1} \) by the following condition:

\[
B_{k+1} = \begin{cases} 
B_k - \frac{B_k s_k s_k^T B_k}{s_k^T B_k s_k} + \frac{y_k y_k^T}{y_k^T s_k}, & \text{if } \frac{y_k^T s_k}{||s_k||^2} \geq \varepsilon ||g_k||^\alpha \\
B_k, & \text{otherwise}
\end{cases}
\]

(44)

where \( \varepsilon \) and \( \alpha \) are positive constants.

The inverse of \( B_{k+1} \) can be obtained by applying the Sherman–Morrison formula:

\[
B_{k+1}^{-1} = B_k^{-1} + \frac{(y_k^T s_k + y_k^T B_k^{-1} y_k)(s_k s_k^T)}{(y_k^T s_k)^2} - \frac{B_k^{-1} y_k s_k s_k^T B_k^{-1} y_k^T}{y_k^T s_k}
\]

(45)

In practical applications, to avoid any matrix inversion, the inverse of Hessian matrix instead of the Hessian matrix itself is approximated. Denoting \( B_k^{-1} \) by \( H_k \), then Eq. (40) and Eq. (45) become:

\[
p_k = -H_k g_k
\]

(46)

and

\[
H_{k+1} = H_k + \frac{(y_k^T s_k + y_k^T H_k y_k)(s_k s_k^T)}{(y_k^T s_k)^2} - \frac{H_k y_k s_k^T + s_k y_k^T H_k}{y_k^T s_k}
\]

(47)

respectively. Accordingly, \( H_{k+1} \) will be determined using the criteria:
In general, the BFGS method with a cautious update is summarized as follows:

**Step 1:** Choose an initial $\beta_0 \in \mathbb{R}^m$, an initial inverse Hessian approximation $H_0 \in \mathbb{R}^{m \times m}$, a convergence tolerance $\kappa > 0$, constants $\rho, \sigma \in (0, 1)$ and $\varepsilon, \alpha > 0$. Let $k := 0$;

**Step 2:** Terminate the iteration if $\|g_k\|^2 < \kappa$;

**Step 3:** Compute search direction $N_k$ using Eq. (46);

**Step 4:** Determine step size $\gamma_k$ that satisfies Eq. (42);

**Step 5:** Compute $\beta_{k+1}$ using Eq. (39);

**Step 6:** Determine $H_{k+1}$ using Eq. (48);

**Step 7:** Let $k := k + 1$ and go to step 2;

In our case of minimizing $\chi^2$, the gradient of $\chi^2$ can be calculated using the equation:

$$\nabla \chi^2 = J^T r$$

(49)

where $J$ is the Jacobian matrix with $J_{ij}$ being:

$$J_{ij} = \frac{\partial r_i}{\partial \beta_j}$$

(50)

$J$ can be analytically solved in our problem, and the solution is given in Appendix A.

Once the parameters $\beta_i$ are optimized, the 1-σ error on $\beta_i$ can be obtained. The asymptotic standard parameter errors $\sigma$ are given by the equation (Clifford, 1973):

$$\sigma = \sqrt{\text{diag}([J^T J]^{-1})}$$

(51)

And the 1-σ error on $\beta_i$ is given by:

$$\sigma(\beta_i) = \sigma_{i\ell}$$

(52)
IV. Results

4.1 Fitting diffusion profiles with mass fraction as concentration

Optimized eigenvector matrix $[P_w]$ and eigenvalues obtained by fitting mass fraction data together with the 1-σ errors are shown in Table 4.1. Generally, eigenvalues are listed in ascending order. All features of the diffusion profiles from the diffusion couple experiments are well reproduced by the optimal parameters (e.g., Figs. 4.2-4.4).

<table>
<thead>
<tr>
<th>Eigenvalues</th>
<th>$\lambda_1$</th>
<th>$\lambda_2$</th>
<th>$\lambda_3$</th>
<th>$\lambda_4$</th>
<th>$\lambda_5$</th>
<th>$\lambda_6$</th>
<th>$\lambda_7$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1260 °C</td>
<td>1.95±0.04</td>
<td>1.98±0.09</td>
<td>3.60±0.11</td>
<td>20.35±0.50</td>
<td>22.16±0.51</td>
<td>27.1±0.44</td>
<td>168.98±1.90</td>
</tr>
<tr>
<td>1350 °C</td>
<td>6.88±0.22</td>
<td>4.72±0.24</td>
<td>13.11±0.38</td>
<td>34.41±0.95</td>
<td>31.47±0.94</td>
<td>47.46±1.03</td>
<td>207.27±3.64</td>
</tr>
<tr>
<td>1500 °C</td>
<td>17.76±0.61</td>
<td>18.5±0.76</td>
<td>37.07±0.88</td>
<td>101.35±1.73</td>
<td>104.2±1.83</td>
<td>147.53±2.41</td>
<td>456.17±9.14</td>
</tr>
</tbody>
</table>

Table 4.1. Eigenvalues at different temperatures and eigenvector matrix $[P_w]$ fitted by mass fraction data.

<table>
<thead>
<tr>
<th>Eigenvectors</th>
<th>$\nu_1$</th>
<th>$\nu_2$</th>
<th>$\nu_3$</th>
<th>$\nu_4$</th>
<th>$\nu_5$</th>
<th>$\nu_6$</th>
<th>$\nu_7$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO$_2$</td>
<td>-1.45</td>
<td>-0.78</td>
<td>-0.59</td>
<td>0.25</td>
<td>-0.38</td>
<td>-0.04</td>
<td>-0.19</td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>0.64±0.04</td>
<td>-0.08±0.09</td>
<td>-0.34±0.03</td>
<td>0.03±0.01</td>
<td>-0.06±0.01</td>
<td>0.02±0.01</td>
<td>-0.01±0.00</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>-0.01±0.14</td>
<td>0.98±0.55</td>
<td>-0.20±0.03</td>
<td>0.01±0.02</td>
<td>-0.09±0.02</td>
<td>-0.03±0.01</td>
<td>-0.1±0.02</td>
</tr>
<tr>
<td>FeO</td>
<td>0.55±0.03</td>
<td>-0.07±0.08</td>
<td>0.64±0.02</td>
<td>-0.56±0.12</td>
<td>0.88±0.1</td>
<td>-0.63±0.06</td>
<td>-0.39±0.07</td>
</tr>
<tr>
<td>MgO</td>
<td>0.28±0.02</td>
<td>-0.11±0.08</td>
<td>0.43±0.02</td>
<td>-0.23±0.11</td>
<td>-0.17±0.09</td>
<td>-0.05±0.02</td>
<td>-0.16±0.03</td>
</tr>
<tr>
<td>CaO</td>
<td>0.38±0.01</td>
<td>0.09±0.03</td>
<td>0.43±0.01</td>
<td>0.76±0.11</td>
<td>-0.37±0.09</td>
<td>0.69±0.05</td>
<td>-0.07±0.01</td>
</tr>
<tr>
<td>Na$_2$O</td>
<td>-0.19±0.01</td>
<td>-0.01±0.01</td>
<td>-0.17±0.01</td>
<td>-0.02±0.02</td>
<td>-0.05±0.02</td>
<td>-0.23±0.01</td>
<td>0.90±0.16</td>
</tr>
<tr>
<td>K$_2$O</td>
<td>-0.19</td>
<td>-0.03</td>
<td>-0.20</td>
<td>-0.25</td>
<td>0.23</td>
<td>0.26</td>
<td>0.02</td>
</tr>
</tbody>
</table>

Note: $\lambda$ is in $\mu$m$^2$/s. The “SiO$_2$” term in each eigenvector was obtained by taking the inverse of the sum of all elements in that eigenvector. In the eigenvector matrix, all positive elements are shown in bold and errors greater than 0.20 are highlighted in red.

The obtained eigenvalues at different temperatures were fitted by the Arrhenius relation:

$$\ln \lambda = \ln \lambda_0 - \frac{E_a}{RT},$$

where $\lambda_0$ is the pre-exponential factor in $\mu$m$^2$/s, $E_a$ is activation energy in kJ/mol, $R$ is the universal gas constant (8.31447 J mol$^{-1}$ K$^{-1}$), and $T$ is temperature in K. The fitted dependence of $\lambda$ on temperature is listed in Table 4.2 and plotted in Fig. 4.1. All eigenvalues roughly follow the Arrhenius relation with temperature, but some eigenvalues are close to each other (e.g., $\lambda_1$ and $\lambda_2$; $\lambda_4$, $\lambda_5$ and $\lambda_6$), a phenomenon defined to be near degeneracy.
by Guo and Zhang (2020). Mathematically, an eigenvalue is said to be degenerate if corresponding eigenspace has dimension greater than one. In this case, any vector in that eigenspace will be an eigenvector corresponding to the eigenvalue. The near degeneracy of our eigenvalues means that our eigenvectors may not be well constrained, and hence more constraints are needed to improve our eigenvector matrix.

**Table 4.2.** Temperature dependence of eigenvalues listed in Table 4.1 and corresponding activation energies.

<table>
<thead>
<tr>
<th>Eigenvalues</th>
<th>Arrhenius relation</th>
<th>Activation Energy (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\lambda_1$</td>
<td>$\ln \lambda_1 = 16.820 - 24552/T$</td>
<td>204</td>
</tr>
<tr>
<td>$\lambda_2$</td>
<td>$\ln \lambda_2 = 17.225 - 25390/T$</td>
<td>211</td>
</tr>
<tr>
<td>$\lambda_3$</td>
<td>$\ln \lambda_3 = 18.337 - 25947/T$</td>
<td>216</td>
</tr>
<tr>
<td>$\lambda_4$</td>
<td>$\ln \lambda_4 = 14.940 - 18366/T$</td>
<td>153</td>
</tr>
<tr>
<td>$\lambda_5$</td>
<td>$\ln \lambda_5 = 14.675 - 17917/T$</td>
<td>149</td>
</tr>
<tr>
<td>$\lambda_6$</td>
<td>$\ln \lambda_6 = 15.884 - 19374/T$</td>
<td>161</td>
</tr>
<tr>
<td>$\lambda_7$</td>
<td>$\ln \lambda_7 = 12.566 - 11522/T$</td>
<td>96</td>
</tr>
</tbody>
</table>

Notes: $\lambda_i$ is in $\mu m^2/s$ and $T$ is temperature in K.

**Figure 4.1.** Temperature dependence of eigenvalues listed in Table 4.1. The straight lines are fits by Arrhenius relation.
Figure 4.2. Diffusion profiles of oxide components in wt% of BS7&8C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure 4.3. Diffusion profiles of oxide components in wt% of BS19&20C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure 4.4. Diffusion profiles of oxide components in wt% of BS11&12A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
4.2 Fitting diffusion profiles with mole fraction as concentration

Optimized eigenvector matrix \([P_X]\) and eigenvalues obtained by fitting mole fraction data together with the 1-σ errors are shown in Table 4.3. Eigenvalues are listed generally in ascending order. All features of the diffusion profiles from the diffusion couple experiments are well reproduced by the optimal parameters (e.g., Figs. 4.6-4.8).

Table 4.3. Eigenvalues at different temperatures and eigenvector matrix \([P_X]\) fitted by mole fraction data.

<table>
<thead>
<tr>
<th>Eigenvalues</th>
<th>(\lambda_1)</th>
<th>(\lambda_2)</th>
<th>(\lambda_3)</th>
<th>(\lambda_4)</th>
<th>(\lambda_5)</th>
<th>(\lambda_6)</th>
<th>(\lambda_7)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1260 °C</td>
<td>1.92±0.08</td>
<td>1.95±0.04</td>
<td>3.57±0.10</td>
<td>19.70±0.46</td>
<td>22.12±0.45</td>
<td>27.30±0.45</td>
<td>168.56±1.86</td>
</tr>
<tr>
<td>1350 °C</td>
<td>4.71±0.21</td>
<td>6.99±0.22</td>
<td>13.57±0.38</td>
<td>35.46±0.94</td>
<td>31.27±0.86</td>
<td>48.00±1.02</td>
<td>205.40±3.44</td>
</tr>
<tr>
<td>1500 °C</td>
<td>18.36±0.68</td>
<td>17.39±0.56</td>
<td>37.28±0.87</td>
<td>102.11±1.64</td>
<td>103.63±1.74</td>
<td>148.44±2.37</td>
<td>459.07±8.42</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Eigenvectors</th>
<th>(v_1)</th>
<th>(v_2)</th>
<th>(v_3)</th>
<th>(v_4)</th>
<th>(v_5)</th>
<th>(v_6)</th>
<th>(v_7)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO(_2)</td>
<td>0.73</td>
<td>-1.33</td>
<td>-0.55</td>
<td>-0.24</td>
<td>0.29</td>
<td>-0.03</td>
<td>-0.33</td>
</tr>
<tr>
<td>TiO(_2)</td>
<td>-0.06±0.06</td>
<td>0.47±0.03</td>
<td>-0.23±0.02</td>
<td>0.02±0.01</td>
<td>-0.04±0.01</td>
<td>0.01±0.00</td>
<td>-0.01±0.01</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td>(0.98\pm0.48)</td>
<td>0.12±0.15</td>
<td>-0.19±0.03</td>
<td>(0.01\pm0.02)</td>
<td>-0.07±0.01</td>
<td>-0.04±0.01</td>
<td>-0.13±0.07</td>
</tr>
<tr>
<td>FeO</td>
<td>-0.07±0.06</td>
<td>0.47±0.02</td>
<td>0.49±0.02</td>
<td>-0.28±0.09</td>
<td>(0.80\pm0.08)</td>
<td>-0.47±0.05</td>
<td>-0.22±0.12</td>
</tr>
<tr>
<td>MgO</td>
<td>-0.15±0.10</td>
<td>0.42±0.04</td>
<td>(0.59\pm0.02)</td>
<td>-0.44±0.13</td>
<td>-0.23±0.10</td>
<td>-0.07±0.03</td>
<td>-0.17±0.09</td>
</tr>
<tr>
<td>CaO</td>
<td>0.07±0.01</td>
<td>0.43±0.01</td>
<td>(0.42\pm0.01)</td>
<td>(0.80\pm0.11)</td>
<td>-0.42±0.08</td>
<td>(0.70\pm0.05)</td>
<td>-0.09±0.05</td>
</tr>
<tr>
<td>Na(_2)O</td>
<td>-0.02±0.01</td>
<td>-0.35±0.01</td>
<td>-0.30±0.01</td>
<td>-0.06±0.03</td>
<td>-0.09±0.02</td>
<td>-0.42±0.02</td>
<td>(0.95\pm0.50)</td>
</tr>
<tr>
<td>K(_2)O</td>
<td>-0.03</td>
<td>-0.24</td>
<td>-0.23</td>
<td>-0.28</td>
<td>(0.34)</td>
<td>(0.32)</td>
<td>0.00</td>
</tr>
</tbody>
</table>

Note: \(\lambda\) is in \(\mu \text{m}^2/\text{s}\). The “SiO\(_2\)” term in each eigenvector was obtained by taking the inverse of the sum of all elements in that eigenvector. In the eigenvector matrix, all positive elements are shown in bold and errors greater than 0.20 are highlighted in red.

As an analogy to Part 4.1, the obtained eigenvalues at different temperatures were also fitted using the Arrhenius relation (see Table 4.4) and then plotted in Fig. 4.5. It can be observed that all eigenvalues listed in Table 4.3 roughly follow the Arrhenius relation with temperature. However, despite using mole fraction as concentration, the near degeneracies of \(\lambda_1\) and \(\lambda_2\) and of \(\lambda_4\), \(\lambda_5\) and \(\lambda_6\) still exist.
Table 4.4. Temperature dependence of eigenvalues listed in Table 4.3 and corresponding activation energies.

<table>
<thead>
<tr>
<th>Eigenvalues</th>
<th>Arrhenius relation</th>
<th>Activation Energy (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ln λ₁</td>
<td>17.337 − 25595/T</td>
<td>213</td>
</tr>
<tr>
<td>ln λ₂</td>
<td>16.650 − 24276/T</td>
<td>202</td>
</tr>
<tr>
<td>ln λ₃</td>
<td>18.420 − 26065/T</td>
<td>217</td>
</tr>
<tr>
<td>ln λ₄</td>
<td>15.180 − 18755/T</td>
<td>156</td>
</tr>
<tr>
<td>ln λ₅</td>
<td>14.647 − 17879/T</td>
<td>149</td>
</tr>
<tr>
<td>ln λ₆</td>
<td>15.880 − 19353/T</td>
<td>161</td>
</tr>
<tr>
<td>ln λ₇</td>
<td>12.634 − 11636/T</td>
<td>97</td>
</tr>
</tbody>
</table>

Notes: λᵢ is in µm²/s and T is temperature in K.

Figure 4.5. Temperature dependence of eigenvalues listed in Table 4.3. The straight lines are fits by Arrhenius relation.
Figure 4.6. Diffusion profiles of oxide components in mole% of BS7&8C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure 4.7. Diffusion profiles of oxide components in mole% of BS19&20C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure 4.8. Diffusion profiles of oxide components in mole% of BS11&12A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
V. Discussion

5.1 Comparison to eigenvector matrices from previous studies

Guo and Zhang (2018, 2020) fit the three sets of diffusion couple data using mass fraction as concentration at different temperatures separately and obtained three similar eigenvector matrices, indicating that the diffusion eigenvectors in basaltic melts do not depend much on temperature. Our fitting results show that one eigenvector matrix with three sets of eigenvalues fit all concentration profiles well, more rigorously demonstrating that the diffusion eigenvectors are invariant with temperature. In addition, Guo and Zhang (2020) have estimated a constant eigenvector matrix (referred to as \([P_0]\) hereafter) by taking the weighted average of eigenvectors at three different temperatures. The \([P_0]\) matrix together with our \([P_w]\) matrix are given in Table 5.1. It is readily seen that two matrices are pretty much similar in terms of \(v_2, v_3, v_5, v_6\) and \(v_7\), but still show apparent differences in \(v_1\) and \(v_4\).

<table>
<thead>
<tr>
<th>([P_0])</th>
<th>(v_1)</th>
<th>(v_2)</th>
<th>(v_3)</th>
<th>(v_4)</th>
<th>(v_5)</th>
<th>(v_6)</th>
<th>(v_7)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiO(_2)</td>
<td>0.76</td>
<td>-0.20</td>
<td>-0.18</td>
<td>0.02</td>
<td>-0.02</td>
<td>-0.02</td>
<td>-0.02</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td>0.18</td>
<td>0.97</td>
<td>-0.47</td>
<td>0.01</td>
<td>-0.15</td>
<td>-0.07</td>
<td>-0.10</td>
</tr>
<tr>
<td>FeO</td>
<td>0.51</td>
<td>0.00</td>
<td>0.66</td>
<td>-0.06</td>
<td>0.86</td>
<td>-0.41</td>
<td>-0.36</td>
</tr>
<tr>
<td>MgO</td>
<td>0.17</td>
<td>-0.03</td>
<td>0.41</td>
<td>0.71</td>
<td>-0.14</td>
<td>-0.32</td>
<td>-0.15</td>
</tr>
<tr>
<td>CaO</td>
<td>0.22</td>
<td>0.12</td>
<td>0.33</td>
<td>-0.70</td>
<td>-0.33</td>
<td>0.79</td>
<td>-0.08</td>
</tr>
<tr>
<td>Na(_2)O</td>
<td>-0.17</td>
<td>-0.04</td>
<td>-0.18</td>
<td>0.04</td>
<td>-0.12</td>
<td>-0.19</td>
<td>0.91</td>
</tr>
<tr>
<td>K(_2)O</td>
<td>-0.13</td>
<td>-0.02</td>
<td>-0.09</td>
<td>0.10</td>
<td>0.32</td>
<td>0.25</td>
<td>0.06</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>([P_w])</th>
<th>(v_1)</th>
<th>(v_2)</th>
<th>(v_3)</th>
<th>(v_4)</th>
<th>(v_5)</th>
<th>(v_6)</th>
<th>(v_7)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiO(_2)</td>
<td>0.64</td>
<td>-0.08</td>
<td>-0.34</td>
<td>0.03</td>
<td>-0.06</td>
<td>0.02</td>
<td>-0.01</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td>-0.01</td>
<td>0.98</td>
<td>-0.20</td>
<td>0.01</td>
<td>-0.09</td>
<td>-0.03</td>
<td>-0.10</td>
</tr>
<tr>
<td>FeO</td>
<td>0.55</td>
<td>-0.07</td>
<td>0.64</td>
<td>-0.56</td>
<td>0.88</td>
<td>-0.63</td>
<td>-0.39</td>
</tr>
<tr>
<td>MgO</td>
<td>0.28</td>
<td>-0.11</td>
<td>0.43</td>
<td>-0.23</td>
<td>-0.17</td>
<td>-0.05</td>
<td>-0.16</td>
</tr>
<tr>
<td>CaO</td>
<td>0.38</td>
<td>0.09</td>
<td>0.43</td>
<td>0.76</td>
<td>-0.37</td>
<td>0.69</td>
<td>-0.07</td>
</tr>
<tr>
<td>Na(_2)O</td>
<td>-0.19</td>
<td>-0.01</td>
<td>-0.17</td>
<td>-0.02</td>
<td>-0.05</td>
<td>-0.23</td>
<td>0.90</td>
</tr>
<tr>
<td>K(_2)O</td>
<td>-0.19</td>
<td>-0.03</td>
<td>-0.20</td>
<td>-0.25</td>
<td>0.23</td>
<td>0.26</td>
<td>0.02</td>
</tr>
</tbody>
</table>

Note: All positive elements are shown in bold.
5.2 Comparison of \( [P_w] \) and \( [P_X] \)

The eigenvector matrix \( [P_w] \) is obtained based on the assumption that the mass density \( \rho \) is roughly constant across the diffusion profiles, whereas the eigenvector matrix \( [P_X] \) is obtained based on the assumption that the single cation molar density \( \rho_i \) is roughly constant across the diffusion profiles. Even though theoretically these two conditions cannot be satisfied simultaneously, we see that the experimental data from Guo and Zhang (2018, 2020) show small compositional variations across the diffusion profiles and hence we are inclined to neglect both density variations. If both conditions roughly hold, it can be seen from Eq. (26) and Eq. (31) that the two eigenvector matrices should satisfy the equation:

$$ [P_X] = \left[M^{-1}\right] [P_w] $$  \hspace{1cm} (53)

where in our case,

$$ [M^N] = \begin{bmatrix} M_{TiO_2} & M_{AlO_{1.5}} & M_{FeO} & M_{Mgo} \\ M_{FeO} & M_{Mgo} & M_{CaO} & M_{NaO_{0.5}} \\ M_{Mgo} & M_{CaO} & M_{Kao_{0.5}} & M_{KO_{0.5}} \end{bmatrix} $$  \hspace{1cm} (54)

Using Eqs. (53)-(54) and the eigenvector matrix \( [P_w] \) listed in Table 4.1, the matrix \( \left[M^{-1}\right] [P_w] \) was calculated. To make it comparable to the eigenvector matrix \( [P_X] \) listed in Table 4.3, each column vector of \( \left[M^{-1}\right] [P_w] \) was normalized so that its 2-norm equals to 1.

The normalized \( \left[M^{-1}\right] [P_w] \), denoted as \( [P^*_X] \), as well as \( [P_X] \) are given in Table 5.2. We tested whether matrices \( [P^*_X] \) and \( [P_X] \) are different by calculating \( T_{ij} = (P^*_{X,ij} - P_{X,ij})/\sqrt{\sigma_{ij}^2 + \sigma_{ij}^2} \). If \( [P^*_X] \) and \( [P_X] \) agree with each other within error, then the absolute values of \( T_{ij} \) would be less
than 2 at 95% probability assuming a standard normal distribution. Our calculation shows that all
\( T_{ij} \) values have an absolute value less than 2, thus \( [P_X^*] \) and \( [P_X] \) are the same within errors.

Nevertheless, it can be clearly seen that some elements in \( [P_X^*] \) and \( [P_X] \) are quite different, e.g.,
the (2,2), (3,4) and (4,4) entries in both matrices. The discrepancy can be attributed to the two
assumptions we made about mass density \( \rho \) and molar density \( \tilde{\rho} \), both of which cannot be
rigorously constant when composition varies across diffusion profiles.

### Table 5.2. Eigenvector matrix \([P_X^*]\) and eigenvector matrix \([P_X]\).

<table>
<thead>
<tr>
<th>([P_X^*])</th>
<th>(v_1)</th>
<th>(v_2)</th>
<th>(v_3)</th>
<th>(v_4)</th>
<th>(v_5)</th>
<th>(v_6)</th>
<th>(v_7)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiO(_2)</td>
<td>-0.05±0.06</td>
<td>0.49±0.03</td>
<td>-0.23±0.02</td>
<td>0.02±0.01</td>
<td>-0.04±0.01</td>
<td>0.01±0.00</td>
<td>0.00±0.00</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td><strong>0.98±0.55</strong></td>
<td>-0.01±0.17</td>
<td>-0.21±0.04</td>
<td>0.02±0.03</td>
<td>-0.11±0.03</td>
<td>-0.03±0.01</td>
<td>-0.07±0.01</td>
</tr>
<tr>
<td>FeO</td>
<td>-0.05±0.06</td>
<td><strong>0.47±0.02</strong></td>
<td>0.49±0.02</td>
<td>-0.45±0.10</td>
<td>0.79±0.09</td>
<td>-0.49±0.05</td>
<td>-0.18±0.03</td>
</tr>
<tr>
<td>MgO</td>
<td>-0.14±0.1</td>
<td><strong>0.42±0.04</strong></td>
<td>0.58±0.02</td>
<td>-0.32±0.15</td>
<td>-0.28±0.14</td>
<td>-0.06±0.03</td>
<td>-0.13±0.02</td>
</tr>
<tr>
<td>CaO</td>
<td><strong>0.09±0.02</strong></td>
<td><strong>0.41±0.01</strong></td>
<td><strong>0.42±0.01</strong></td>
<td><strong>0.78±0.11</strong></td>
<td>-0.42±0.10</td>
<td><strong>0.70±0.05</strong></td>
<td>-0.04±0.01</td>
</tr>
<tr>
<td>Na(_2)O</td>
<td>-0.01±0.02</td>
<td>-0.38±0.01</td>
<td>-0.30±0.01</td>
<td>-0.04±0.04</td>
<td>-0.10±0.03</td>
<td>-0.42±0.02</td>
<td><strong>0.97±0.17</strong></td>
</tr>
<tr>
<td>K(_2)O</td>
<td>-0.03</td>
<td>-0.25</td>
<td>-0.23</td>
<td>-0.31</td>
<td><strong>0.32</strong></td>
<td><strong>0.31</strong></td>
<td><strong>0.01</strong></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>([P_X])</th>
<th>(v_1)</th>
<th>(v_2)</th>
<th>(v_3)</th>
<th>(v_4)</th>
<th>(v_5)</th>
<th>(v_6)</th>
<th>(v_7)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiO(_2)</td>
<td>-0.06±0.06</td>
<td><strong>0.47±0.03</strong></td>
<td>-0.23±0.02</td>
<td><strong>0.02±0.01</strong></td>
<td>-0.04±0.01</td>
<td><strong>0.01±0.00</strong></td>
<td>-0.01±0.01</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td><strong>0.98±0.48</strong></td>
<td><strong>0.12±0.15</strong></td>
<td>-0.19±0.03</td>
<td><strong>0.01±0.02</strong></td>
<td>-0.07±0.01</td>
<td>-0.04±0.01</td>
<td>-0.13±0.07</td>
</tr>
<tr>
<td>FeO</td>
<td>-0.07±0.06</td>
<td><strong>0.47±0.02</strong></td>
<td><strong>0.49±0.02</strong></td>
<td>-0.28±0.09</td>
<td><strong>0.80±0.08</strong></td>
<td>-0.47±0.05</td>
<td>-0.22±0.12</td>
</tr>
<tr>
<td>MgO</td>
<td>-0.15±0.10</td>
<td><strong>0.42±0.04</strong></td>
<td><strong>0.59±0.02</strong></td>
<td>-0.44±0.13</td>
<td>-0.23±0.10</td>
<td>-0.07±0.03</td>
<td>-0.17±0.09</td>
</tr>
<tr>
<td>CaO</td>
<td><strong>0.07±0.01</strong></td>
<td><strong>0.43±0.01</strong></td>
<td><strong>0.42±0.01</strong></td>
<td><strong>0.80±0.11</strong></td>
<td>-0.42±0.08</td>
<td><strong>0.70±0.05</strong></td>
<td>-0.09±0.05</td>
</tr>
<tr>
<td>Na(_2)O</td>
<td>-0.02±0.01</td>
<td>-0.35±0.01</td>
<td>-0.30±0.01</td>
<td>-0.06±0.03</td>
<td>-0.09±0.02</td>
<td>-0.42±0.02</td>
<td><strong>0.95±0.50</strong></td>
</tr>
<tr>
<td>K(_2)O</td>
<td>-0.03</td>
<td>-0.24</td>
<td>-0.23</td>
<td>-0.28</td>
<td><strong>0.34</strong></td>
<td><strong>0.32</strong></td>
<td>0.00</td>
</tr>
</tbody>
</table>

Note: All positive elements are shown in bold and errors greater than 0.20 are highlighted in red.

### 5.3 Verification of the universality of eigenvector matrix

To test our hypothesis that eigenvector matrix is invariant with composition in natural
silicate melts, we transformed the oxide concentrations into eigen-component “concentrations”
and plotted the profiles of eigen-components as a function of distance using data from earlier
diffusion experiments (Zhang et al., 1989; Chen and Zhang, 2008, 2009; Yu et al., 2016; Guo
and Zhang, 2016; González-Garcia et al., 2017; Yu et al., 2019; Guo and Zhang, 2018, 2020;
Wang et al., 2020). In total, there are 171 experiments and 1197 concentration profiles. The two eigenvector matrices \([P_w]\) and \([P_x]\) were tested separately. Profiles of eigen-components using mass fraction as concentration were calculated using Eq. (33) to test the universality of \([P_w]\), whereas profiles of eigen-components using mole fraction as concentration were calculated using Eq. (34) to test the universality of \([P_x]\). In the former case, about 3% (35 out of 1197) profiles are non-monotonic (Table 5.3), of which 23% are from crystal dissolution experiments in andesite (Zhang et al., 1989), 20% are from plagioclase dissolution experiments in basalt (Yu et al., 2016) and 29% are from diffusion couple experiments in haplo-basalt (Guo and Zhang, 2016). In the latter case, roughly 2% (28 out of 1197) profiles are non-monotonic (Table 5.4), of which 32% are from crystal dissolution experiments in andesite (Zhang et al., 1989), 25% are from diffusion couple experiments in haplo-basalt (Guo and Zhang, 2016) and 18% are from shoshonite-rhyolite diffusion couple experiments (González-García et al., 2017). In both cases, all diffusion profiles of quartz dissolution experiments in basalt and rhyolite are monotonic, even though the compositional variations are large across the profiles. Overall, most profiles (e.g., Figs. 5.1-5.6) are monotonic and show no oblivious uphill diffusion, supporting our hypothesis that the matrix is independent of melt composition.

Table 5.3. Summary of the 35 \([P_w]\)-converted concentration profiles of eigen-components that show obvious uphill diffusion, along with the corresponding experimental conditions. The remaining 1162 profiles of eigen-components are monotonic.

<table>
<thead>
<tr>
<th>Exp</th>
<th>Melt comp</th>
<th>Type</th>
<th>(Z_1)</th>
<th>(Z_2)</th>
<th>(Z_3)</th>
<th>(Z_4)</th>
<th>(Z_5)</th>
<th>(Z_6)</th>
<th>(Z_7)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zhang1989_Exp#225a</td>
<td>Andesite</td>
<td>DiDisAn</td>
<td>×</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Zhang1989_Exp#236RUa</td>
<td>Andesite</td>
<td>RuDisAn</td>
<td>×</td>
<td>×</td>
<td>×</td>
<td>×</td>
<td>×</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Zhang1989_Exp#259PLa</td>
<td>Andesite</td>
<td>PiDisAn</td>
<td>×</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chen2008_Exp#44b</td>
<td>JDF Basalt</td>
<td>OlDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chen2009_Exp#5c</td>
<td>JDF Basalt</td>
<td>CpxDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yu2016_PiDisBa202d</td>
<td>JDF Basalt</td>
<td>PiDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yu2016_PiDisBa210d</td>
<td>JDF Basalt</td>
<td>PiDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yu2016_PiDisBa211d</td>
<td>JDF Basalt</td>
<td>PiDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yu2016_PiDisBa214d</td>
<td>JDF Basalt</td>
<td>PiDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yu2016_PiDisBa216d</td>
<td>JDF Basalt</td>
<td>PiDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yu2016_PiDisBa233d</td>
<td>JDF Basalt</td>
<td>PiDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yu2016_PiDisBa230d</td>
<td>JDF Basalt</td>
<td>PiDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Guo2016_HB2&amp;4Ae</td>
<td>Haplobasalt</td>
<td>DC</td>
<td>×</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Guo2016_HB3&amp;4Ae</td>
<td>Haplobasalt</td>
<td>DC</td>
<td>×</td>
<td>×</td>
<td>×</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Guo2016_HB5&amp;6Ae</td>
<td>Haplobasalt</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Guo2016_HB7&amp;8Be</td>
<td>Haplobasalt</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
</tr>
</tbody>
</table>
Table 5.4. Summary of the 28 $[P_X]$-converted concentration profiles of eigen-components that show obvious uphill diffusion, along with the corresponding experimental conditions. The remaining 1169 profiles of eigen-components are monotonic.

<table>
<thead>
<tr>
<th>Exp</th>
<th>Base comp</th>
<th>Type</th>
<th>$Z_1$</th>
<th>$Z_2$</th>
<th>$Z_3$</th>
<th>$Z_4$</th>
<th>$Z_5$</th>
<th>$Z_6$</th>
<th>$Z_7$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zhang1989_Exp#220</td>
<td>Andesite</td>
<td>ODisAn</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
</tr>
<tr>
<td>Zhang1989_Exp#225</td>
<td>Andesite</td>
<td>DiDisAn</td>
<td>×</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
</tr>
<tr>
<td>Zhang1989_Exp#234</td>
<td>Andesite</td>
<td>QzDisAn</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Zhang1989_Exp#259PL</td>
<td>Andesite</td>
<td>RuDisAn</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Yu2016_PDisBa202</td>
<td>JDF Basalt</td>
<td>PDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Yu2016_PDisBa211</td>
<td>JDF Basalt</td>
<td>PDisBa</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Guo2016_HB3&amp;4A</td>
<td>Haplobasalt</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Guo2016_HB5&amp;6A</td>
<td>Haplobasalt</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Guo2016_HB7&amp;8B</td>
<td>Haplobasalt</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Guo2016_HB15&amp;16A</td>
<td>Haplobasalt</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Guo2020_BS11&amp;12C</td>
<td>Basalt</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Guo2020_BS17&amp;18C</td>
<td>Basalt</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>González2017_P050-H0-4</td>
<td>Shoshonite &amp; Rhyolite</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>González2017_P300-H0-4</td>
<td>Shoshonite &amp; Rhyolite</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>González2017_P300-H2-4</td>
<td>Shoshonite &amp; Rhyolite</td>
<td>DC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>×</td>
<td>×</td>
</tr>
</tbody>
</table>

Note: see notes in Table 5.3.

5.4 Diffusion mechanism in natural silicate melts

Eigenvector matrix characterizes the diffusion mechanism in natural silicate melts.

Previous studies interpreted the diffusion mechanism by directly checking the terms in each eigenvector, with the assumption that the oxide components corresponding to the terms with large absolute values contribute more during diffusion and the oxide components corresponding to the terms with different signs diffuse against each other (e.g., Guo and Zhang, 2018, 2020).

However, we note in Eqs. (33)- (34) that it is the terms in the inverse of the eigenvector matrix that comprise the “stoichiometric” coefficients in the formula of eigen-components expressed by oxide components. Instead, the terms in the eigenvector matrix are basically the “stoichiometric”
coefficients to express oxide components by eigen-components. Furthermore, the way of looking at the components in eigenvectors only works for interpreting the exchange mechanism among eigen-components instead of oxide components. As a result, to truly interpret the exchange mechanism among oxide components, it is necessary to first calculate the inverse of the eigenvector matrix. Furthermore, to better understand the diffusion exchange mechanism in natural silicate melts, the eigenvector matrix $[P_X]$ instead of $[P_w]$ should be used.

Table 5.5. The inverse of the eigenvector matrix $[P_X]$.  

<table>
<thead>
<tr>
<th></th>
<th>SiO$_2$</th>
<th>TiO$_2$</th>
<th>AlO$_{1.5}$</th>
<th>FeO</th>
<th>MgO</th>
<th>CaO</th>
<th>NaO$_{0.5}$</th>
<th>KO$_{0.5}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Z_1$</td>
<td>-1.46</td>
<td>-0.42</td>
<td>1.02</td>
<td>0.13</td>
<td>0.11</td>
<td>0.20</td>
<td>0.21</td>
<td>0.21</td>
</tr>
<tr>
<td>$Z_2$</td>
<td>-2.72</td>
<td>1.45</td>
<td>0.14</td>
<td>0.30</td>
<td>0.29</td>
<td>0.28</td>
<td>0.18</td>
<td>0.09</td>
</tr>
<tr>
<td>$Z_3$</td>
<td>-0.54</td>
<td>-1.26</td>
<td>0.02</td>
<td>0.42</td>
<td>0.61</td>
<td>0.52</td>
<td>0.25</td>
<td>-0.02</td>
</tr>
<tr>
<td>$Z_4$</td>
<td>1.84</td>
<td>-0.18</td>
<td>-0.22</td>
<td>0.27</td>
<td>-1.08</td>
<td>0.42</td>
<td>-0.12</td>
<td>-0.93</td>
</tr>
<tr>
<td>$Z_5$</td>
<td>-1.32</td>
<td>-0.09</td>
<td>-0.03</td>
<td>0.89</td>
<td>-0.61</td>
<td>0.26</td>
<td>0.12</td>
<td>0.78</td>
</tr>
<tr>
<td>$Z_6$</td>
<td>-2.66</td>
<td>0.07</td>
<td>0.05</td>
<td>-0.18</td>
<td>0.37</td>
<td>0.69</td>
<td>0.09</td>
<td>1.57</td>
</tr>
<tr>
<td>$Z_7$</td>
<td>-3.46</td>
<td>0.13</td>
<td>0.08</td>
<td>0.27</td>
<td>0.34</td>
<td>0.63</td>
<td>1.25</td>
<td>0.75</td>
</tr>
</tbody>
</table>

Note: All positive elements are shown in bold. The “SiO$_2$” term in each row was obtained by the sum of all elements in that row multiplied by -1.

The inverse of $[P_X]$ was calculated and shown in Table 5.5. We see that the $Z_1$ (one of the two slowest diffusing eigen-components) is due to the exchange between SiO$_2$ + TiO$_2$ and Al$_2$O$_3$ + everything else; $Z_2$ (the other one of the two slowest diffusing eigen-components) is due to the exchange between SiO$_2$ and TiO$_2$ + everything else; $Z_3$ (the third slowest diffusing eigen-component) is mostly due to the exchange between TiO$_2$ + SiO$_2$ and divalent cations; $Z_4$ and $Z_5$ (the fourth and the third fastest diffusing eigen-components) are mostly due to the exchange between SiO$_2$ and MgO + K$_2$O, and the exchange between SiO$_2$ + MgO and FeO + K$_2$O, respectively; $Z_6$ (the second fastest diffusing eigen-component) are mostly due to the exchange between SiO$_2$ and MgO + CaO + K$_2$O; $Z_7$ (the fastest diffusing eigen-component) are mostly due to the exchange between SiO$_2$ and all alkalis.
Figure 5.1. \([P_m]\)-converted concentration profiles of eigen-components of Exp#227 (Zhang et al., 1989). Exp#227 is a forsterite dissolution experiment in andesite.
Figure 5.2. $[P_w]_c$-converted concentration profiles of eigen-components of Exp#40 (Chen and Zhang, 2008). Exp#40 is an olivine dissolution experiment in basalt.
Figure 5.3. $[P_w]$-converted concentration profiles of eigen-components of QzDisRh106 (Yu et al., 2019). QzDisRh106 is a quartz dissolution experiment in rhyolite.
Figure 5.4. $[P_x]$-converted concentration profiles of eigen-components of Exp#227 (Zhang et al., 1989). Exp#227 is a forsterite dissolution experiment in andesite.
Figure 5.5. $[P_x]$-converted concentration profiles of eigen-components of Exp#40 (Chen and Zhang, 2008). Exp#40 is an olivine dissolution experiment in basalt.
Figure 5.6. $[P_X]$-converted concentration profiles of eigen-components of QzDisRh106 (Yu et al., 2019). QzDisRh106 is a quartz dissolution experiment in rhyolite.
VI. Summary and Implications

In this study, we hypothesize that there exists a universal diffusion eigenvector matrix in natural silicate melts. The eigenvector matrix and eigenvalue matrices were obtained by simultaneously fitting diffusion profiles from 27 diffusion couple experiments from previous studies (Guo and Zhang, 2018, 2020). Using both mass fraction as concentration and mole fraction as concentration, we obtained two eigenvector matrices, $[P_w]$ and $[P_X]$, respectively. Both eigenvector matrices were used to examine the “concentration” profiles of eigen components. Most profiles of eigen-components from numerous diffusion experiments including diffusion-couple and mineral-dissolution experiments are monotonic and show no obvious uphill diffusion, supporting that the matrix of eigenvectors is invariable with composition. Finally, diffusion mechanism were interpreted from the eigenvector matrix $[P_X]$.

With the universal eigenvector matrix, investigations, and applications of multicomponent diffusion in natural silicate melts would be greatly simplified in different aspects. Theoretically, the diffusion mechanism in natural silicate melts, which is interpreted from the eigenvector matrix, would also be independent of melt composition. And the eigen-components will also be invariable with melt composition. Experimentally, to accurately extract a diffusivity matrix at selected melt composition typically need at least N-1 experiments for an N-component system (e.g., Trial and Spera, 1994). With an accurate eigenvector matrix, however, only a couple experiment may be needed to extract N-1 unknown eigenvalues. In addition, since the eigen-components are universal, characterization of the compositional dependence of eigenvalues would be viable.

Multicomponent diffusion is also key to understanding magma mixing processes, in which two or more magmas come into contact and form a new magma with a different composition. In the absence of convection or other fluid dynamic processes, multicomponent diffusion can be the
exclusive mechanism to account for the complicated compositional variations at the interface of two melts (Chakraborty et al., 1995b). With the universal eigenvector matrix, modeling and prediction of melt composition at the diffusive interface will be simplified. Here we give an example to show compositional variations along the diffusive interface of dacite and basalt at 1500 °C. We did the calculation using our universal eigenvector matrix \( [P_\xi] \), and assuming eigenvalues are 2 times smaller than our optimal eigenvalues at 1500 °C listed in Table 4.3. The calculated profiles are shown in Fig. 6.1. \( \text{TiO}_2, \text{Al}_2\text{O}_3, \text{FeO}, \text{Na}_2\text{O} \) and \( \text{K}_2\text{O} \) show clear uphill diffusion features. Specifically, \( \text{TiO}_2, \text{Al}_2\text{O}_3, \) and \( \text{FeO} \) are depleted at \( x \approx -400 \, \mu m \), but enriched at \( x \approx 400 \, \mu m \), while \( \text{Na}_2\text{O} \) and \( \text{K}_2\text{O} \) exhibit complementary patterns. The melt compositions at \( x \approx \pm 400 \, \mu m \) may be captured as inclusions in other melts or early formed crystals. If these melt inclusions reach the surface and are collected as research samples, however, the chemical compositions will be difficult to explain exclusively by a binary mixing model. Therefore, in future studies of magma mixing, the kinetic control of multicomponent diffusion must be considered to explain the potential composition anomalies in melt inclusions.
Figure 6.1. Calculated concentration profiles resulting from magma mixing between dacite and basalt at 1500 °C, using the eigenvector matrix \([P_\lambda]\) and assuming eigenvalues are 2 times smaller than our optimal eigenvalues at 1500 °C listed in Table 4.3.
References


Appendix A

Derivation of an explicit expression of Jacobian matrix

The Jacobian matrix, which is defined in Eq. (50), is a key parameter in the fitting process. It determines both the search direction (see Eq. (40)) and the 1-σ error of optimal fitting parameters (see Eq. (51)). The Jacobian matrix can be easily obtained using a “universal” numerical method. One disadvantage of the method is that the calculated matrix may not be accurate. Thus, if there is a high demand for data accuracy and the function is not too complicated, one may try to derive an explicit expression of Jacobian matrix. We see that the Jacobian matrix in our case is essentially the partial derivatives of oxide concentrations with respect to eigenvector matrix and eigenvalues. Thus, once such partial derivatives are explicitly expressed, the Jacobian matrix can be accurately obtained.

In an $N$-component melt system, choose the $N^{th}$ oxide component as the dependent component. Let $n = N - 1$ and $w_i$ be the mass fraction of the $i^{th}$ oxide component. Then according to mass conservation, we have

$$w_N = 100\% - \sum_{i=1}^{n} w_i \quad (A1)$$

Define the concentration vector $\mathbf{w}$ as follows:

$$\mathbf{w} = \begin{pmatrix} w_1 \\ w_2 \\ \vdots \\ w_n \end{pmatrix} \quad (A2)$$

Assuming a constant diffusivity matrix, the analytical solution to an infinite diffusion couple is given by:

$$\mathbf{w} = \frac{\mathbf{w}_2 + \mathbf{w}_1}{2} + [P] \times [E] \times [P^{-1}] \times \frac{\mathbf{w}_2 - \mathbf{w}_1}{2} \quad (A3)$$
where $\mathbf{w}_1$ and $\mathbf{w}_2$ are initial melt concentration vectors at $x < 0$ and $x > 0$, $[P]$ is eigenvector matrix and $[E]$ is a diagonal matrix:

$$
[E] = \begin{bmatrix}
\text{erf} \left( \frac{x-x_0}{\sqrt{4e^{\theta_1}t}} \right) \\
\vdots \\
\text{erf} \left( \frac{x-x_0}{\sqrt{4e^{\theta_n}t}} \right)
\end{bmatrix}
$$

(A4)

Define $\theta$ vector as follows:

$$
\theta = \begin{pmatrix}
\theta_1 \\
\vdots \\
\theta_n
\end{pmatrix}
$$

(A5)

Now let’s derive the partial derivative of $\mathbf{w}$ with respect to $[P]$ and $\theta$.

Define $Y$ vector as follows:

$$
Y = \begin{pmatrix}
\frac{x-x_0}{\sqrt{4t}} e^{-\frac{\theta_1}{2}} \\
\vdots \\
\frac{x-x_0}{\sqrt{4t}} e^{-\frac{\theta_n}{2}}
\end{pmatrix}
= \frac{x-x_0}{\sqrt{4t}} e^{-\frac{\theta}{2}}
$$

(A6)

Then $[E]$ matrix can be expressed by $Y$:

$$
[E] = \text{diag} \left( \text{erf} \left( \frac{x-x_0}{\sqrt{4t}} e^{-\frac{\theta_1}{2}} \right), \ldots, \text{erf} \left( \frac{x-x_0}{\sqrt{4t}} e^{-\frac{\theta_n}{2}} \right) \right) = \text{diag} \left( \text{erf} \left( Y \right) \right)
$$

(A7)

where diag is a function so that diag($v$) returns a square diagonal matrix with the elements of vector $v$ on the main diagonal and diag([$A$]) returns a column vector of the main diagonal elements of matrix $[A]$.

In Eq. (A3), take the total differential of $\mathbf{w}$:
\[ \mathbf{d} \mathbf{w} = \mathbf{d} \left[ P \right] \times [E] \times [P^{-1}] \times \frac{\mathbf{w}_2 - \mathbf{w}_1}{2} + [P] \times \mathbf{d} \left[ E \right] \times [P^{-1}] \times \frac{\mathbf{w}_2 - \mathbf{w}_1}{2} \]

\[ + [P] \times [E] \times \mathbf{d} \left[ P^{-1} \right] \times \frac{\mathbf{w}_2 - \mathbf{w}_1}{2} \]  

(A8)

Our next step is to express the total differential of \( \mathbf{w} \) by the total differential of \([P]\) and \(\theta\). There are three terms on the right-hand side (RHS) of Eq. (A8), but only the first term is a desired form. Now let's calculate \( \mathbf{d} \left[ E \right] \) in the second term and \( \mathbf{d} \left[ P^{-1} \right] \) in the third term on the RHS of Eq. (A8). In the following part, we use the symbol \(\odot\) to denote the entry wise product of two vectors (or two matrices) of the same size and use the symbol \(\otimes\) to denote Kronecker product of two matrices.

In Eq. (A7), take the total differential of \([E]\):

\[ \mathbf{d} \left[ E \right] = \mathbf{d} \text{ diag}(\text{erf}(\mathbf{Y})) = \text{diag}(\mathbf{d} \text{ erf}(\mathbf{Y})) \]

\[ = \text{diag} \left( \frac{2}{\sqrt{\pi}} e^{-\mathbf{Y} \odot \mathbf{Y}} \odot \mathbf{d} \mathbf{Y} \right) \]

\[ = \text{diag} \left( \frac{2}{\sqrt{\pi}} e^{-\mathbf{Y} \odot \mathbf{Y}} \odot \mathbf{d} \left( \frac{x - x_0}{\sqrt{4t}} e^{-\frac{\theta}{2}} \right) \right) \]

\[ = \text{diag} \left( \frac{2}{\sqrt{\pi}} e^{-\mathbf{Y} \odot \mathbf{Y}} \odot \left( \frac{x - x_0}{\sqrt{4t}} \ast \left( -\frac{1}{2} \ast e^{-\frac{\theta}{2}} \right) \ldots \mathbf{d} \mathbf{\theta} \right) \right) \]

\[ = \text{diag} \left( \frac{-\left( x - x_0 \right)}{\sqrt{4\pi t}} e^{-\mathbf{Y} \odot \mathbf{Y}} \odot e^{-\frac{\theta}{2}} \odot \mathbf{d} \mathbf{\theta} \right) \]

\[ = \text{diag} \left( \frac{-\left( x - x_0 \right)}{\sqrt{4\pi t}} e^{-\mathbf{Y} \odot \mathbf{Y}} \odot e^{-\frac{\theta}{2}} \right) \odot \mathbf{d} \left( \text{diag} \mathbf{\theta} \right) \]

\[ = [dE] \odot \mathbf{d} \left( \text{diag} \mathbf{\theta} \right) \]  

(A9)

where the matrix \([dE]\) is defined as:

\[ [dE] = \text{diag} \left( \frac{-\left( x - x_0 \right)}{\sqrt{4\pi t}} e^{-\mathbf{Y} \odot \mathbf{Y}} \odot e^{-\frac{\theta}{2}} \right) \]

(A10)

Therefore, the second term on the RHS of Eq. (A8) can be expressed as follows:
in the third term on the RHS of Eq. (A8) can be calculated as follows:

$$ d \left[ P^{-1} \right] = - \left[ P^{-1} \right] \times d \left[ P \right] \times \left[ P^{-1} \right] $$

(A12)

Plugging Eq. (A11) and Eq. (A12) into Eq. (A8), leads to:

$$ d w = d \left[ P \right] \times \left[ E \right] \times \left[ P^{-1} \right] \times \frac{w_2 - w_1}{2} $$

$$ + \left[ P \right] \times \left( \left[ dE \right] \odot d \left( \text{diag} \ \theta \right) \right) \times \left[ P^{-1} \right] \times \frac{w_2 - w_1}{2} $$

$$ - \left[ P \right] \times \left[ E \right] \times \left[ P^{-1} \right] \times d \left[ P \right] \times \left[ P^{-1} \right] \times \frac{w_2 - w_1}{2} $$

(A13)

Now we have expressed the total differential of $w$ by the total differentials of $[P]$ and $\text{diag} \ \theta$. To make sure that the partial derivative of $w$ with respect to $[P]$ and $\text{diag} \ \theta$ is a 2-dimensional matrix, we need to convert $[P]$ and $\text{diag} \ \theta$ into 1-dimensional column vectors.

Taking the vectorization of first term on the RHS of Eq. (A13), leads to

$$ \text{vec} \left( d \left[ P \right] \times \left[ E \right] \times \left[ P^{-1} \right] \times \frac{w_2 - w_1}{2} \right) $$

$$ = \left( \left[ E \right] \times \left[ P^{-1} \right] \times \frac{w_2 - w_1}{2} \right)^T \otimes [I_n] \times \text{vec} \left( d \left[ P \right] \right) $$

(A14)

where $[I_n]$ is an $n$ square identity matrix.

Taking the vectorization of second term on the RHS of Eq. (A13), leads to

$$ \text{vec} \left( \left[ P \right] \times \left( \left[ dE \right] \odot d \left( \text{diag} \ \theta \right) \right) \times \left[ P^{-1} \right] \times \frac{w_2 - w_1}{2} \right) $$

$$ = \left( \left[ P^{-1} \right] \times \frac{w_2 - w_1}{2} \right)^T \otimes [P] \times \text{vec} \left( \left[ dE \right] \odot d \left( \text{diag} \ \theta \right) \right) $$

$$ = \left( \left[ P^{-1} \right] \times \frac{w_2 - w_1}{2} \right)^T \otimes [P] \times \text{diag} \left( \text{vec} \left( \left[ dE \right] \right) \right) \times \text{vec} \left( d \left( \text{diag} \ \theta \right) \right) $$

(A15)

And taking the vectorization of last term on the RHS of Eq. (A13), we have
\[
\text{vec} \left( -[P] \times [E] \times [P^{-1}] \times d \ [P] \times [P^{-1}] \times \frac{W_2 - W_1}{2} \right)
\]
\[
= - \left( \left( [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes ([P] \times [E] \times [P^{-1}]) \right) \times \text{vec}(d \ [P])
\]  
(A16)

According to Eqs. (A14) - (A16), the vectorization of \( d \ w \) is as following:

\[
\text{vec}(d \ w)
\]
\[
= \left\{ \left( [E] \times [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes [I_n] \right\} - \left( \left( [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes ([P] \times [E] \times [P^{-1}]) \right) \times \text{vec}(d \ [P])
\]
\[
+ \left( \left( [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes [P] \right) \times \text{diag}(\text{vec}([dE])) \times \text{vec}(d \text{ (diag } \theta))
\]  
(A17)

Therefore, the partial derivative of \( w \) with respect to \([P]\) can be expressed as:

\[
[w_p] = \frac{\partial w}{\partial [P]} = \left\{ \left( [E] \times [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes [I_n] \right\} - \left( \left( [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes ([P] \times [E] \times [P^{-1}]) \right)^T
\]
\[
= \left( [E] \times [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes [I_n] \right\} - \left( \left( [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes ([P] \times [E] \times [P^{-1}]) \right)^T
\]  
(A18)

And the partial derivative of \( w \) with respect to [diag \( \theta \)] can be expressed as:

\[
[w_{\text{diag } \theta}] = \frac{\partial w}{\partial [\text{diag } \theta]} = \left\{ \left( \left( [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes [P] \right) \times \text{diag}(\text{vec}([dE])) \right\}^T
\]
\[
= \text{diag}(\text{vec}([dE])) \times \left( \left( [P^{-1}] \times \frac{W_2 - W_1}{2} \right)^T \otimes [P^T] \right)
\]  
(A19)

According to Eq. (A1), we have:

\[
d \ w_N = - \sum_{i=1}^{n} d \ w_i
\]  
(A20)

Therefore, the partial derivatives of \( w_N \) with respect to \([P]\) and [diag \( \theta \)] are as follows:

\[
\frac{\partial w_N}{\partial [P]} = - \sum_{i=1}^{n} [w_p](; i)
\]  
(A21)

and
\[ \frac{\partial w_N}{\partial [\text{diag } \theta]} = -\sum_i^n [w_{\text{diag } \theta}](i, i) \]  

where \([w_P](i, i)\) is the \(i^{th}\) column of matrix \([w_P]\) and \([w_{\text{diag } \theta}](i, i)\) is the \(i^{th}\) column of matrix \([w_{\text{diag } \theta}]\).

If we use the symbol \(\bigodot\) to express the element-wise division of two vectors of the same size, then the vector of residual difference can be expressed by the formula:

\[ r = (w_0 - w) \bigodot \sigma \]  

where \(w_0\) is the vector of measured concentrations of \(n\) independent oxide components, \(\sigma\) is the 1-\(\sigma\) error on \(w_0\).

Then the partial derivatives of \(r\) with respect to \([P]\) and \([\text{diag } \theta]\) are given by:

\[ [r_P] = \frac{\partial r}{\partial [P]} = -\frac{\partial w}{\partial [P]} \bigodot \sigma = -[w_P] \bigodot \sigma \]  

and

\[ [r_{\text{diag } \theta}] = \frac{\partial r}{\partial [\text{diag } \theta]} = -\frac{\partial w}{\partial [\text{diag } \theta]} \bigodot \sigma = -[w_{\text{diag } \theta}] \bigodot \sigma \]  

where \([w_P]\) and \([w_{\text{diag } \theta}]\) can be obtained by Eq. (A18) and Eq. (A19), respectively.

Finally, the partial derivative of the residual difference \(r_N\) of the \(N^{th}\) components are given by:

\[ \frac{\partial r_N}{\partial [P]} = \frac{\partial \left(\frac{w_{0,N} - w_N}{\sigma_N}\right)}{\partial [P]} = -\frac{1}{\sigma_N} \frac{\partial w_N}{\partial [P]} \]  

and

\[ \frac{\partial r_N}{\partial [\text{diag } \theta]} = \frac{\partial \left(\frac{w_{0,N} - w_N}{\sigma_N}\right)}{\partial [\text{diag } \theta]} = -\frac{1}{\sigma_N} \frac{\partial w_N}{\partial [\text{diag } \theta]} \]
where $\frac{\partial w_N}{\partial [p]}$ and $\frac{\partial w_N}{\partial [\text{diag } \theta]}$ can be obtained by Eq. (A21) and Eq. (A22).

Once all derivatives of all residual differences are calculated, according to Eq. (50), the Jacobian matrix can be obtained.
Appendix B

MATLAB codes for fitting diffusion profiles

B1. Main program “BFGS_Main.m”

%% this program uses the BFGS method to obtain eigenvectors and eigenvalues by simultaneously fitting diffusion profiles.

% Input parameters are stored in the files: "Initial_beta.xlsx"
% Input parameters should be a 10×7 matrix, with the last 7 rows
% being eigenvector matrix and the first 3 rows being eigenvalues.

% Once the program ends, the reduced chi-square is stored as the variable
% "S_DF", fitted parameters are stored as "trans_beta" and the 1-sigma errors
% are sorted as "error_trans_beta".

close all;
clear all;
start_time = clock();

%% ----------------------------define a structure-----------------------------
data.temp= 1260; %1260, 1350,1500
data.type= 1;% 1:diffusion couple(DC); 2:mineral dissolution(MD)
data.np = 100; % number of points for each exp
data.dist = 1;
% for DC: data.dist is the location of the interface ; for MD: data.dist is
% the growth thickness of the melt
data.time = 1000;% exp duration
data.boundary = [];%boundary condition; size:[8,2]
data.x = [];% x-coordinate;[np,1]
data.w = [];% concentration;[np,8]
data.w_calc = [];% predicted concentration;[np,8];
data.error=[];% error of concentration measured by EMPA

%% ------------------------initialize----------------------------------------
T = [1260,1350,1500];%exp temp

% exp duration at different temp
t1 = [1826.0, 1809.6, 1325.5, 1269.2, 950.8, 579.5, 740.0, 577.4, 599.9,...
     335.4, 1981.2, 395.3]; % exp duration at 1260

% exp duration at different temp
t2 = [1492.4, 1243.2, 899.0, 974.4, 563.9, 393.6, 522.0, 335.7, 346.3,...
     298, 1163, 601, 742, 1611, 395, 3087, 121, 2138]; % at 1350

% exp duration at different temp
t3 = [276.1, 223.6, 247.8, 213.7, 158.5, 157.1, 154.8, 184.9, 221.4]; % at
1500

Np1 = [112,121,128,140,97,146,123,128,120,146,54,102];% num of points of each exp

Np2 = [116,84,100,109,105,101,64,122,123,191,226,183,191,152,136,166,91,160];% number of points of exp at 1260

Np3 = [152,136,166,91,160];% number of points at 1350
Np3 = [92,124,116,118,120,141,131,141,166];
% number of points at 1500
Np=[Np1,Np2,Np3];

Ndm = [12,18,9]; % number of exp at 1260, 1350 and 1500
threshold = [0,12,30]; % no actual meaning, just for reading data

% x0 and L of each exp
% for diffusion couple exp, we have already adjusted x0 to be 0
A1 = [0.158, 0.12, 0.295];  
% constant alpha in mineral dissolution exp at 1260
L1 = 2*A1.*sqrt(t1(10:12));
% growth thickness of the melt in mineral dissolution exp at 1260
A2 = [0.5174, 0.5174, 0.5174, 1.1961, 0.9985, 0.9985, 0.9985, 0.9985, 0.9985];
%constant alpha at 1350
L2 = 2*A2.*sqrt(t2(10:18));  
% growth thickness of the melt at 1350
L = [zeros(1,9),L1,zeros(1,9),L2,zeros(1,9)];

Nexp = 39; % total number of exp

%read data
for i=1:3
    T_str = string(T(i));
    realdata = xlsread(strcat(T_str,"_RealData_8Comp.xlsx"));
    realerror = xlsread(strcat(T_str,"_RealError_8Comp.xlsx"));
    realboundary = xlsread(strcat(T_str,"_RealBoundary_8Comp.xlsx"));
    for j=1:Ndm(i)
        j1 = j+threshold(i);
        data(j1).temp = T(i);
        if j<10
            data(j1).type = 1;
        else
            data(j1).type = 2;
        end
        data(j1).np= Np(j1);
        data(j1).dist = L(j1);
        data(j1).time= t(j1);
        data(j1).boundary = realboundary(1:8,(3*j-2):(3*j-1));
        data(j1).x = realdata(1:Np(j1),12*j-11);
        data(j1).w = realdata(1:Np(j1),(12*j-10):(12*j-3));
        data(j1).error=realerror(:,j);
    end
end

% read initial beta
read_b = xlsread("Initial_beta.xlsx");

% ---------------------Choose data to fit-------------------------------
data_d = [data(1:9),data(13:21),data(31:39)];  %only DC data
% data_d = [data(10:12),data(22:30)];%only MD data
% data_d = data; %all data
[~,m]=size(data_d); % number of exp involved in fitting
Nc = 7; %7 independent components;

Npoint = 0;
for i = 1:m
    Npoint = Npoint + data_d(i).np;
end

% DF = Nc*Npoint - 49; % only one temperature included
% DF = Nc*Npoint - 56; % two temperatures included
DF = Nc*Npoint - 63; % three temperatures included

error = [];
for i = 1:m
    Np_i = data_d(i).np;
    error_i = kron(ones(Np_i,1),data_d(i).error);
    error = [error;error_i];
end

P = read_b(4:10,1:7);% matrix of eigenvector of D
Beta1 = read_b(1,1:7)';% initial log of eigenvalue at 1260 C
Beta2 = read_b(2,1:7)';% initial log of eigenvalue at 1350 C
Beta3 = read_b(3,1:7)';% initial log of eigenvalue at 1500 C

% Normalize eigenvectors
Num_Max = [1,2,3,5,3,5,6];
% the positions of the largest terms in each eigenvector; only for DC
% Num_Max = [2,2,3,3,3,5,6];
% the positions of the largest terms in each eigenvector; all data
for i = 1:7
    Max = beta(7*i-7+Num_Max(i));
    beta(7*i-6:7*i) = beta(7*i-6:7*i) / norm(beta(7*i-6:7*i)) * (Max/abs(Max));
end

[w0,~] = size(beta);

w0 = []; % initial concentration
for i = 1:m
    transform_w = reshape((data_d(i).w)',[],1);
    w0 = [w0;transform_w];
end
w = con(data_d,beta); %calculate concentration
r = (w-w0)./error; % calculate residue difference
S = r'*r; % sum of square error
S_DF = S/DF/2;  % reduced chi-square  
J = calculate_J(data_d,beta);  % calculate Jacobian matrix  
g = 2*J'*r;  % gradient of S  
H = eye(70);  % H matrix  

x = [x;n_loop1];  
y1 = [y1;S_DF];  
y2 = [y2;n_loop2];  

yyaxis left  
plot(x,y1,'-b');  
xlabel('num of loop1');  
ylabel('reduced chi-square');  
yyaxis right  
plot(x,y2,'-*r');  
ylabel('num of loop2');  
drawnow;  

max1 = 800;  
max2 = 50;  
sigma = 0.5;  
% control parameter used in Armijo-type line search  
rho = 0.5;  
% control parameter used in Armijo-type line search  
n_loop1 = 1;  

while n_loop1 < max1 && norm(g)/DF/2 > 1E-6  
% when n_loop1 exceeds max1 or ||S_DF|| > 1E-6, iteration ends  

p = -H*g;  
% the BFGS direction obtained by solving the linear equation: p=-H*g  
step = 1;  
n_loop2 = 0;  

% search for proper step using Armijo-type line search.  
while n_loop2 < max2  
betal1 = beta + step*p;  
w1 = con(data_d,betal1);  
r1 = (w0-w1)./error;  
S1 = r1'*r1;  
l = S + sigma*step*g'*p;  % parameter used in Armijo-type line search  

if S1 < l, the step is OK, otherwise, reduce the step by a factor of rho  
if S1 < 1  
    break;  
end  
step = rho*step;  
n_loop2 = n_loop2+1;  
end  

if n_loop2==max2  
    fprintf("n_loop2 = %d \n",max2);  
    break;  
end  

s = betal1 - beta;  % increase in beta  
% update beta  
beta = betal1;  
% normalize eigenvectors  
for i =1:7  
    Max = beta( 7*i - 7 + Num_Max(i));  
end
beta(7*i-6:7*i) = beta(7*i-6:7*i) / norm(beta(7*i-6:7*i)) * (Max/abs(Max));
end

% update w,r,S,S_DF,J and y
w = w1;
r = r1;
S = S1;
S_DF = S/DF/2;
J = calculate_J(data_d,beta);
y = 2*J'*r - g;

% update H matrix to guarantee the positive definiteness of H
if norm(g)>1
  if (y'*s)/(s'*s)>= 10^-6*norm(g)^0.01
    H = H + (y'*s + y'*H*y)*(s*s')/(y'*s)^2 - (H*y*s'+s*y'*H)/(y'*s);
  end
else
  if (y'*s)/(s'*s)>= 10^-6*norm(g)^3
    H = H + (y'*s + y'*H*y)*(s*s')/(y'*s)^2 - (H*y*s'+s*y'*H)/(y'*s);
  end
end

% update g
g = 2*J'*r;

x = [x;n_loop1];
y1 = [y1;S_DF];
y2 = [y2;n_loop2];
plot(x,y1,'-b');
xlabel('num of loop1');
ylabel('reduced chi-square');
plot(x,y2,'-*r');
ylabel('num of loop2');
drawnow;
n_loop1 = n_loop1 +1;
end

% Normalize eigenvectors
for i = 1:7
  Max = beta(7*i-7+Num_Max(i));
  beta(7*i-6:7*i) = beta(7*i-6:7*i) / norm(beta(7*i-6:7*i)) * (Max/abs(Max));
end

% ------------------------end of BFGS method------------------------

transform_w = reshape(w,8,[]); j=0;

% store calculated concentration
for i=1:m
  data_d(i).w_calc = transform_w(j+1:j+data_d(i).np,:);
  j = j + data_d(i).np;
end
% using obtained beta to predict concentration profiles of all exp
w_predict = con(data,beta);
transform_w = reshape(w_predict,8,[]);'
j=0;
for i=1:Nexp
    data(i).w_calc = transform_w(j+1:j+data(i).np,:); 
    j = j + data(i).np;
end

% calculate eigenvalues
%trans_beta is a 10 by 7 matrix.
% First row: log of eigenvalues at 1260
% Second row: log of eigenvalues at 1350
% Third row: log of eigenvalues at 1500
% The rest: matrix of eigenvector

trans_beta = zeros(10,7);  % change dimensions of beta
trans_beta(1:3,:) = reshape(beta(50:70) ,7,3);  % calculate lambda of each exp
trans_beta(4:10,:) = reshape(beta(1:49),7,7);

% sequence eigenvalues using bubble sort
for i = 1:6
    for j = 1:7-i
        if trans_beta(1,j)>trans_beta(1,j+1)
            store = trans_beta(:,j+1);
            trans_beta(:,j+1)=trans_beta(:,j);
            trans_beta(:,j)= store;
        end
    end
end

% calculate D matrix at each temperature
D1 = trans_beta(4:10,:)*diag(exp(trans_beta(1,:)))/trans_beta(4:10,:); %1260
D2 = trans_beta(4:10,:)*diag(exp(trans_beta(2,:)))/trans_beta(4:10,:); %1350
D3 = trans_beta(4:10,:)*diag(exp(trans_beta(3,:)))/trans_beta(4:10,:); %1500

% calculate error of trans_beta
beta(1:49) = reshape(trans_beta(4:10,:),49,1);
beta(50:70) = reshape(trans_beta(1:3,:),',21,1);
J7 = calculate_J7(data_d,beta);
A = J7'*J7;
inv_A = (diag(inv(A))).^0.5;

error_beta = zeros(70,1);
for i = 0:6
    error_beta(7*i+1:7*i+6)= inv_A(6*i+1:6*i+6);
end
error_beta(50:70) = inv_A(43:63);

error_trans_beta = zeros(10,7);
error_trans_beta(1:3,:) = reshape( error_beta(50:70) , 7,3);'
error_trans_beta(4:10,:) = reshape( error_beta(1:49) , 7,7);
elapsed_time = etime(clock(),start_time);
B2. Subroutine “con.m”

%% Calculate concentration using assumed eigenvector matrix and eigenvalues.

function ww = con(data_d,beta)
    [~,m]=size(data_d);
P = reshape(beta(1:49),7,7);
ww = [];
for i=1:m
    w1 = data_d(i).boundary(2:8,1);
    w2 = data_d(i).boundary(2:8,2);
    %for DC, w1 = w_left, w2 = w_right
    %for MD, w1 = w_interface, w2 = w_initial
    x0 = data_d(i).dist;
t = data_d(i).time;

    if data_d(i).temp == 1260
        Beta = beta(50:56);
    elseif data_d(i).temp == 1350
        Beta = beta(57:63);
    else
        Beta = beta(64:70);
    end

    if data_d(i).type==1
        for j=1:data_d(i).np
            x = data_d(i).x(j);
            Y = (x-x0)*exp(-Beta/2)/(sqrt(4*t));
            E = diag( erf(Y) );
            ww1 = (w2 + w1)/2 +P * E *P^-1*(w2-w1)/2;
            %concentration of independent components
            ww2 = 100-sum(ww1,"all");
            %concentration of the dependent component,i.e.,SiO2
            ww = [ww;ww2;ww1];
        end
    elseif data_d(i).type==2
        for j=1:data_d(i).np
            x = data_d(i).x(j);
            Y = (x-x0)*exp(-Beta/2)/(sqrt(4*t));
            Y0 = -x0*exp(-Beta/2)/(sqrt(4*t));
            E = diag( erfc(Y)./erfc(Y0) );
            ww1 = w2 +P * E *P^-1*(w1-w2);
            %concentration of independent components
            ww2 = 100-sum(ww1,"all");
            %concentration of the dependent component,i.e.,SiO2
            ww = [ww;ww2;ww1];
        end
    end
end
end
B3. Subroutine “calculate_J.m"

% Calculate and return the derivates of r with respect to P (matrix of eigenvector) and beta (log of eigenvalue)

function J=calculate_J(data_d,beta)
    [~,m]=size(data_d); %number of experiments included
    P = reshape(beta(1:49),7,7); % matrix of eigenvector
    num = 0;
    n_start = 0;
    n_end = 0;
    JT=[];
    
    for i=1:m
        w1 = data_d(i).boundary(2:8,1);
        w2 = data_d(i).boundary(2:8,2);
        %for DC, w1 = w_left, w2 = w_right
        %for MD, w1 = w_interface, w2 = w_initial
        x0 = data_d(i).dist;
        t = data_d(i).time;
        error1 = data_d(i).error(2:8);
        error2 = data_d(i).error(1);
        if data_d(i).temp == 1260
            num = 0;
            Beta = beta(50:56);
        elseif data_d(i).temp == 1350
            num = 1;
            Beta = beta(57:63);
        else
            num = 2;
            Beta = beta(64:70);
        end
        n_start = num*7+1;
        n_end = (num+1)*7;
    
        if data_d(i).type==1 %for diffusion couple
            for j=1:data_d(i).np
                x= data_d(i).x(j);
                Y= (x-x0)*exp(-Beta/2)/(sqrt(4*t));
                E = diag( erf(Y) ); % Eq.(7) in "derivation1.pdf"
                dE = diag( -(x-x0)/sqrt(4*pi*t) * exp(-Y.^2).*exp(-Beta/2) );
                wp = kron( (E *P^(-1)*(w2-w1)/2, eye(7))-...
                    kron((P^(-1)*(w2-w1)/2), (P*E*P^(-1))');
                % wp is the partial derivative of concentration of
                % independent components with respect to P
                wBeta = diag(reshape(dE,49,1))... *kron((P^(-1)*(w2-w1)/2),P');
                % wBeta is the partial derivative of concentration of
                % independent components with respect to Beta
                rpl1 = -wp/diag(error1);
            end
        end

    end

    JT = JT;

end

% partial derivative of residue error of independent
% components with respect to P
rBeta1= -wBeta/diag(error1);
% partial derivative of residue error of independent
% components with respect to Beta
rp2 = sum(wp,2)/error2;
% partial derivative of residue error of the dependent
% components with respect to P
rBeta2 = sum(wBeta,2)/error2;
% partial derivative of residue error of the dependent
% components with respect to Beta
rp = [rp2,rp1];
% partial derivative of residue error of all
% components with respect to P
rBeta3=[rBeta2,rBeta1];%49*8
% partial derivative of residue error of all
% components with respect to Beta
rBeta=zeros(21,8);
for k=1:8
  rBeta(n_start:n_end,k)= diag(reshape(rBeta3(:,k),7,7));
end
JT =[JT,[rp;rBeta]]; end

elseif data_d(i).type==2  % for mineral dissolution
  for j=1:data_d(i).np
    x= data_d(i).x(j);
    Y= (x-x0)*exp(-Beta/2)/(sqrt(4*t));
    Y0 = -x0*exp(-Beta/2)/(sqrt(4*t));
    E = erfc(Y);
    E0 = erfc(Y0);
    E_ratio = diag (E./E0);
    dE = (x-x0)/(sqrt(4*pi*t))*exp(-Y.*Y).*exp(-Beta/2);
    dE0 = -x0/(sqrt(4*pi*t))*exp(-Y0.*Y0).*exp(-Beta/2);
    dE_ratio = diag( ( dE.*E0 - dE0.*E )./(E0.^2) );
    wp = kron( ( E_ratio*P^-1*(w1-w2)), eye(7)) -...
      kron( (P^-1*(w1-w2)), (P*E_ratio*P^-1)*' ) ;
    % wp is the partial derivative of concentration of
    % independent components with respect to P
    wBeta = diag(reshape(dE_ratio,49,1))...
      *kron((P^-1*(w1-w2)),P');
    % wBeta is the partial derivative of concentration of
    % independent components with respect to Beta
rp1 = -wp/diag(error1);
% partial derivative of residue error of independent
% components with respect with to P

rBeta1= -wBeta/diag(error1);
% partial derivative of residue error of independent
% components with respect with to Beta

rp2 = sum(wp,2)/error2;
% partial derivative of residue error of the dependent
% components with respect with to P

rBeta2 = sum(wBeta,2)/error2;
% partial derivative of residue error of the dependent
% components with respect with to Beta

rp = [rp2,rp1];
% partial derivative of residue error of all
% components with respect with to Beta

rBeta3=[rBeta2,rBeta1];%49*8
% partial derivative of residue error of all
% components with respect with to Beta

rBeta=zeros(21,8);
for k=1:8
    rBeta(n_start:n_end,k)= diag(reshape(rBeta3(:,k),7,7));
end
JT =[JT,[rp;rBeta]];
B4. Subroutine “calculate_J7.m”

% Calculate the derivatives of \( r \) with respect to \( P \) (matrix of eigenvector) and \( \beta \).

% J7 differs from J in that J7 doesn't return the derivatives of \( r \) with respect to the 7th terms of each eigenvector. Therefore, J7 is only used to calculate the error while assuming that the errors of the 7th terms of each eigenvector is zero.

```matlab
function J=calculate_J7(data_d,beta)
    [~,m]=size(data_d);  % number of experiments included
    P = reshape(beta(1:49),7,7);  % matrix of eigenvector
    num = 0;
    n_start = 0;
    n_end = 0;
    JT=[];
    for i=1:m
        w1 = data_d(i).boundary(2:8,1);
        w2 = data_d(i).boundary(2:8,2);
        % for DC, w1 = w_left, w2 = w_right
        % for MD, w1 = w_interface, w2 = w_initial
        x0 = data_d(i).dist;
        t = data_d(i).time;
        error1 = data_d(i).error(2:8);
        error2 = data_d(i).error(1);
        if data_d(i).temp == 1260
            num = 0;
            Beta = beta(50:56);
        elseif data_d(i).temp == 1350
            num = 1;
            Beta = beta(57:63);
        else
            num = 2;
            Beta = beta(64:70);
        end
        n_start = num*7+1;
        n_end = (num+1)*7;
        if data_d(i).type==1  % for diffusion couple
            for j=1:data_d(i).np
                x= data_d(i).x(j);
                Y= (x-x0)*exp(-Beta/2)/(sqrt(4*pi*t));
                E = diag( erf(Y) );
                dE = diag( -(x-x0)/sqrt(4*pi*t) * exp(-Y.*Y).*exp(-Beta/2) );
                wp = kron( (E *P^(-1)*(w2-w1)/2), eye(7))-...
                    kron((P^(-1)*(w2-w1)/2),(P*E*P^(-1)));
                % wp is the partial derivative of concentration
                % of independent components with respect to P
                wBeta = diag(reshape(dE,49,1))...
            end
        end
    end
```
\*kron((P^{-1}*(w2-w1)/2),P');
\% wBeta is the partial derivative of concentration of
\% independent components with respect to Beta

rp1 = -wp/diag(error1);
\% partial derivative of residue error of independent
\% components with respect to P

rBeta1 = -wBeta/diag(error1);
\% partial derivative of residue error of independent
\% components with respect to Beta

rp2 = sum(wp,2)/error2;
\% partial derivative of residue error of the dependent
\% components with respect to P

rBeta2 = sum(wBeta,2)/error2;
\% partial derivative of residue error of the dependent
\% components with respect to Beta

rp = [rp2,rp1];
\% partial derivative of residue error of all
\% components with respect to P

rBeta3 = [rBeta2,rBeta1]; % 49*8
\% partial derivative of residue error of all
\% components with respect to Beta

rBeta = zeros(21,8);
for k=1:8
    rBeta(n_start:n_end,k) = diag(reshape(rBeta3(:,k),7,7));
end
JT = [JT,[rp;rBeta]]; % end

elseif data_d(i).type==2  \% for mineral dissolution

for j=1:data_d(i).np
    x= data_d(i).x(j);
    Y= (x-x0)*exp(-Beta/2)/(sqrt(4*pi*t));
    Y0 = -x0*exp(-Beta/2)/(sqrt(4*pi*t));

    E = erfc(Y);
    E0 = erfc(Y0);
    E_ratio = diag (E./E0);

    dE = (x-x0)/(sqrt(4*pi*t))\*exp(-Y.*Y).\*exp(-Beta/2);
    dE0 = -x0/(sqrt(4*pi*t))\*exp(-Y0.*Y0).\*exp(-Beta/2);

    dE_ratio = diag( ( dE\*E0 - dE0\*E )./(E0.^2) );

    wp = kron( (E_ratio\*P^{-1}*(w1-w2)), eye(7))-...
        kron((P^{-1}*(w1-w2)),(P\*E_ratio\*P^{-1}));
\% wp is the partial derivative of concentration of
\% independent components with respect to P
wBeta = diag(reshape(dE_ratio,49,1))*kron((P^-1*(w1-w2)),P');

% wBeta is the partial derivative of concentration of independent components with respect to Beta

rp1 = -wp/diag(error1);
% partial derivative of residue error of independent components with respect with P

rBeta1= -wBeta/diag(error1);
% partial derivative of residue error of independent components with respect with Beta

rp2 = sum(wp,2)/error2;
% partial derivative of residue error of the dependent components with respect with P

rBeta2 = sum(wBeta,2)/error2;
% partial derivative of residue error of the dependent components with respect with Beta

rp = [rp2,rp1];
% partial derivative of residue error of all components with respect with P

rBeta3=[rBeta2,rBeta1];%49*8
% partial derivative of residue error of all components with respect with Beta

rBeta=zeros(21,8);
for k=1:8
    rBeta(n_start:n_end,k)= diag(reshape(rBeta3(:,k),7,7));
end

JT =[JT,[rp;rBeta]];
Appendix C

Fits for 27 diffusion couple experiments from Guo and Zhang (2018, 2020)

The diffusion profiles of oxide components obtained from 27 diffusion couple experiments conducted by Guo and Zhang (2018, 2020) were fitted twice using two different concentration units, namely mass fraction, and mole fraction. The fits in the former case are shown in Appendix C1, and the fits in the latter case are shown in Appendix C2.
C1. Fits of diffusion profiles using mass fraction as concentration

Figure C1.1. Diffusion profiles of oxide components in wt% of BS1&2C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.2. Diffusion profiles of oxide components in wt% of BS3&4C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.3. Diffusion profiles of oxide components in wt% of BS5&6C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.4. Diffusion profiles of oxide components in wt% of BS7&8C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.5. Diffusion profiles of oxide components in wt% of BS9&10C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.6. Diffusion profiles of oxide components in wt% of BS11&12C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.7. Diffusion profiles of oxide components in wt% of BS13&14C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.8. Diffusion profiles of oxide components in wt% of BS17&18C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.9. Diffusion profiles of oxide components in wt% of BS19&20C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.10. Diffusion profiles of oxide components in wt% of BS1&2A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.11. Diffusion profiles of oxide components in wt% of BS3&4A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.12. Diffusion profiles of oxide components in wt% of BS5&6A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.13. Diffusion profiles of oxide components in wt% of BS7&8A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.14. Diffusion profiles of oxide components in wt% of BS9&10A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.15. Diffusion profiles of oxide components in wt% of BS11&12C (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.16. Diffusion profiles of oxide components in wt% of BS13&14A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.17. Diffusion profiles of oxide components in wt% of BS17&18A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.18. Diffusion profiles of oxide components in wt% of BS19&20C (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.19. Diffusion profiles of oxide components in wt% of BS1&2B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.20. Diffusion profiles of oxide components in wt% of BS3&4B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.21. Diffusion profiles of oxide components in wt% of BS5&6B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.22. Diffusion profiles of oxide components in wt% of BS7&8B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.23. Diffusion profiles of oxide components in wt% of BS9&10B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.24. Diffusion profiles of oxide components in wt% of BS11&12B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.25. Diffusion profiles of oxide components in wt% of BS13&14B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.26. Diffusion profiles of oxide components in wt% of BS17&18B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
Figure C1.27. Diffusion profiles of oxide components in wt% of BS19&20B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.1.
C2. Fits of diffusion profiles using mole fraction as concentration

Figure C2.1. Diffusion profiles of oxide components in mole% of BS1&2C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.2. Diffusion profiles of oxide components in mole\% of BS3&4C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.3. Diffusion profiles of oxide components in mole% of BS5&6C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.4. Diffusion profiles of oxide components in mole% of BS7&8C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.5. Diffusion profiles of oxide components in mole% of BS9&10C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.6. Diffusion profiles of oxide components in mole% of BS11&12C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.7. Diffusion profiles of oxide components in mole% of BS13&14C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.8. Diffusion profiles of oxide components in mole% of BS17&18C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.9. Diffusion profiles of oxide components in mole% of BS19&20C (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.10. Diffusion profiles of oxide components in mole% of BS1&2A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.11. Diffusion profiles of oxide components in mole% of BS3&4A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.12. Diffusion profiles of oxide components in mole% of BS5&6A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.13. Diffusion profiles of oxide components in mole% of BS7&8A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.14. Diffusion profiles of oxide components in mole% of BS9&10A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.15. Diffusion profiles of oxide components in mole% of BS11&12A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.16. Diffusion profiles of oxide components in mole% of BS13&14A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.17. Diffusion profiles of oxide components in mole% of BS17&18A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.18. Diffusion profiles of oxide components in mole% of BS19&20A (Guo and Zhang, 2018) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.19. Diffusion profiles of oxide components in mole% of BS1&2B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.20. Diffusion profiles of oxide components in mole% of BS3&4B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.21. Diffusion profiles of oxide components in mole% of BS5&6B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.22. Diffusion profiles of oxide components in mole% of BS7&8B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.23. Diffusion profiles of oxide components in mole% of BS9&10B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.24. Diffusion profiles of oxide components in mole% of BS11&12B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.25. Diffusion profiles of oxide components in mole% of BS13&14B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.26. Diffusion profiles of oxide components in mole% of BS17&18B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Figure C2.27. Diffusion profiles of oxide components in mole% of BS19&20B (Guo and Zhang, 2020) with fits. The dash curves are fits by the parameters in Table 4.3.
Appendix D

Concentration profiles of oxide components and eigen-components

To facilitate observation, concentration profiles of oxide components in weight percent (wt%), oxide components in mole percent (mole%), $[P_w]$-converted eigen-components and $[P_X]$-converted eigen-components of the same experiment are plotted together. The concentration profiles of 171 experiments, conducted by Zhang et al. (1989), Chen and Zhang (2008, 2009), Yu et al. (2016, 2019), Guo and Zhang (2016, 2018, 2020), González-García et al. (2017), and Wang et al. (2020), are shown in Figs. D1-D171.
Figure D1. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Zhang1989_Exp#212, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D2. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Zhang1989_Exp#216, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D3. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_s]$-converted eigen-components (upper right panel), and $[P_s]$-converted eigen-components (lower right panel) of Zhang1989_Exp#219, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D4. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Zhang1989 Exp#220, which is an olivine dissolution experiment in andesite (Zhang et al., 1989). Note that the duration of the experiment is unknown. Therefore, the x-coordinates were calculated based on $x/2\sqrt{t}$, assuming a value of 1s for t.
Figure D5. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#222, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D6. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_a]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#223, which is a diopside dissolution experiment in andesite (Zhang et al., 1989).
Figure D7. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_\omega]$-converted eigen-components (upper right panel), and $[P_\omega]$-converted eigen-components (lower right panel) of Zhang1989_Exp#225, which is a diopside dissolution experiment in andesite (Zhang et al., 1989).
Figure D8. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_3]$-converted eigen-components (upper right panel), and $[P_3]$-converted eigen-components (lower right panel) of Zhang1989_Exp#226, which is a forsterite dissolution experiment in andesite (Zhang et al., 1989).
Figure D9. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_\text{w}]$-converted eigen-components (upper right panel), and $[P_\text{x}]$-converted eigen-components (lower right panel) of Zhang1989_Exp#227, which is a forsterite dissolution experiment in andesite (Zhang et al., 1989).
Figure D10. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#228FO, which is a forsterite dissolution experiment in andesite (Zhang et al., 1989).
Figure D11. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Zhang1989_Exp#228OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D12. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Zhang1989_Exp#229, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D13. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Zhang1989_Exp#231DI, which is a diopside dissolution experiment in andesite (Zhang et al., 1989).
Figure D14. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_s]\)-converted eigen-components (lower right panel) of Zhang1989_Exp#231OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D15. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#231SP, which is a spinel dissolution experiment in andesite (Zhang et al., 1989).
Figure D16. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_p]$-converted eigen-components (upper right panel), and $[P_p]$-converted eigen-components (lower right panel) of Zhang1989_Exp#234, which is a quartz dissolution experiment in andesite (Zhang et al., 1989).
Figure D17. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#235OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D18. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_w]$-converted eigen-components (lower right panel) of Zhang1989_Exp#236OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D19. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_c]$-converted eigen-components (upper right panel), and $[P_c]$-converted eigen-components (lower right panel) of Zhang1989_Exp#236RU, which is a rutile dissolution experiment in andesite (Zhang et al., 1989).
Figure D20. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_s]$-converted eigen-components (upper right panel), and $[P_s]$-converted eigen-components (lower right panel) of Zhang1989_Exp#239MID OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D21. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#242, which is a diopside dissolution experiment in andesite (Zhang et al., 1989).
Figure D22. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Zhang1989_Exp#246, which is an unknow crystal dissolution experiment in andesite (Zhang et al., 1989).
Figure D23. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#255DI, which is a diopside dissolution experiment in andesite (Zhang et al., 1989).
Figure D24. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_B]$-converted eigen-components (upper right panel), and $[P_B]$-converted eigen-components (lower right panel) of Zhang1989_Exp#255OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D25. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \( [\mathcal{P}_w] \)-converted eigen-components (upper right panel), and \( [\mathcal{P}_X] \)-converted eigen-components (lower right panel) of Zhang1989_Exp#256, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D26. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Zhang1989_Exp#259AUG, which is an augite dissolution experiment in andesite (Zhang et al., 1989).
Figure D27. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_{\text{s}}]$-converted eigen-components (upper right panel), and $[P_{\text{x}}]$-converted eigen-components (lower right panel) of Zhang1989_Exp#259PL, which is a plagioclase dissolution experiment in andesite (Zhang et al., 1989).
Figure D28. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#260OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D29. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Zhang1989_Exp#260SP, which is a spinel dissolution experiment in andesite (Zhang et al., 1989).
Figure D30. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_2]-\)converted eigen-components (upper right panel), and \([P_3]-\)converted eigen-components (lower right panel) of Zhang1989_Exp#262ENS, which is an enstatite dissolution experiment in andesite (Zhang et al., 1989).
Figure D31. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#262OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D32. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#263OL, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D33. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#263SP, which is a spinel dissolution experiment in andesite (Zhang et al., 1989).
Figure D34. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Zhang1989_Exp#265, which is an olivine dissolution experiment in andesite (Zhang et al., 1989).
Figure D35. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P]$-converted eigen-components (upper right panel), and $[Px]$-converted eigen-components (lower right panel) of Chen2008_Exp#15, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D36. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#16, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D37. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_o]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Chen2008_Exp#18, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D38. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Chen2008_Exp#20, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D39. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]-\)converted eigen-components (upper right panel), and \([P_x]-\)converted eigen-components (lower right panel) of Chen2008_Exp#21, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D40. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#22, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D41. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [Pw]-converted eigen-components (upper right panel), and [Px]-converted eigen-components (lower right panel) of Chen2008_Exp#23, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D42. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#24, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D43. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_X]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Chen2008_Exp#25, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D44. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Chen2008_Exp#26, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D45. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Chen2008_Exp#29, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D46. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_e]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#33, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D47. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_s]$-converted eigen-components (lower right panel) of Chen2008_Exp#34, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D48. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P]$-converted eigen-components (upper right panel), and $[PX]$-converted eigen-components (lower right panel) of Chen2008_Exp#35, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D49. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#37, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D50. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_0]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#38, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D51. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Chen2008_Exp#39, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D52. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [Pw]-converted eigen-components (upper right panel), and [PX]-converted eigen-components (lower right panel) of Chen2008_Exp#40, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D53. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#41, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D54. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#43, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D55. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_s]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2008_Exp#44, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D56. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [Pw]-converted eigen-components (upper right panel), and [Px]-converted eigen-components (lower right panel) of Chen2008_Exp#45, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).

176
Figure D57. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Chen2008_Exp#46, which is an olivine dissolution experiment in basalt (Chen and Zhang, 2008).
Figure D58. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P]$-converted eigen-components (upper right panel), and $[P]_X$-converted eigen-components (lower right panel) of Chen2009_Exp#1, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D59. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2009_Exp#5, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D60. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2009_Exp#6, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D61. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2009_Exp#7, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D62. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Chen2009_Exp#8, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D63. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[Pw]$-converted eigen-components (upper right panel), and $[Pw]$-converted eigen-components (lower right panel) of Chen2009_Exp#11, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D64. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]-\)converted eigen-components (upper right panel), and \([P_x]-\)converted eigen-components (lower right panel) of Chen2009_Exp#12, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D65. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2009_Exp14, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D66. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_b]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Chen2009_Exp#15, which is a clinopyroxene dissolution experiment in basalt (Chen and Zhang, 2009).
Figure D67. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2016_Exp#201, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D68. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#202, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D69. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]-\)converted eigen-components (upper right panel), and \([P_X]-\)converted eigen-components (lower right panel) of Yu2016_Exp#203, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D70. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#205, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D71. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [$P_w$]-converted eigen-components (upper right panel), and [$P_X$]-converted eigen-components (lower right panel) of Yu2016_Exp#207, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D72. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [P<sub>w</sub>]-converted eigen-components (upper right panel), and [P<sub>x</sub>]-converted eigen-components (lower right panel) of Yu2016_Exp#208, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D73. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2016_Exp#209, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D74. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#221, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D75. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_\text{w}]$-converted eigen-components (upper right panel), and $[P_\text{x}]$-converted eigen-components (lower right panel) of Yu2016_Exp#222, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D76. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2016_Exp#223, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D77. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_{\text{w}}]$-converted eigen-components (upper right panel), and $[P_{\text{x}}]$-converted eigen-components (lower right panel) of Yu2016_Exp#227, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D78. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#210, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D79. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#211, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
**Figure D80.** Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_x]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Yu2016_Exp#212, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D81. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#213, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D82. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2016_Exp#214, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D83. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2016_Exp#215, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D84. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#216, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D85. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2016_Exp#228, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D86. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#233, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D87. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[Pw]$-converted eigen-components (upper right panel), and $[Px]$-converted eigen-components (lower right panel) of Yu2016_Exp#230, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D88. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_t]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#301, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D89. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2016_Exp#302, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D90. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2016_Exp#304, which is a plagioclase dissolution experiment in basalt (Yu et al., 2016).
Figure D91. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2019_QzDisBa#101, which is a quartz dissolution experiment in basalt (Yu et al., 2019).
Figure D92. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2019_QzDisBa#102, which is a quartz dissolution experiment in basalt (Yu et al., 2019).
Figure D93. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2019_QzDisBa#103, which is a quartz dissolution experiment in basalt (Yu et al., 2019).
Figure D94. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P\omega]$-converted eigen-components (upper right panel), and $[P\omega]$-converted eigen-components (lower right panel) of Yu2019_QzDisBa#104, which is a quartz dissolution experiment in basalt (Yu et al., 2019).
Figure D95. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2019_QzDisBa#107, which is a quartz dissolution experiment in basalt (Yu et al., 2019).
Figure D96. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]-converted\) eigen-components (upper right panel), and \([P_X]-converted\) eigen-components (lower right panel) of Yu2019_QzDisBa#110, which is a quartz dissolution experiment in basalt \(\text{(Yu et al., 2019).}\)
Figure D97. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), [Pw]-converted eigen-components (upper right panel), and [Px]-converted eigen-components (lower right panel) of Yu2019_QzDisBa#111, which is a quartz dissolution experiment in basalt (Yu et al., 2019).
Figure D98. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), [$P_w$]-converted eigen-components (upper right panel), and [$P_X$]-converted eigen-components (lower right panel) of Yu2019_QzDisRh#103, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D99. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2019_QzDisRh#111, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D100. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2019_QzDisRh#112, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D101. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Yu2019_QzDisRh#115, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D102. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [$P_\omega$]-converted eigen-components (upper right panel), and [$P_\omega$]-converted eigen-components (lower right panel) of Yu2019_QzDisRh#201, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D103. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [Pw]-converted eigen-components (upper right panel), and [Pw]-converted eigen-components (lower right panel) of Yu2019_QzDisRh#203, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D104. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2019_QzDisRh#105, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D105. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Yu2019_QzDisRh#113, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D106. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [$P_w$]-converted eigen-components (upper right panel), and [$P_X$]-converted eigen-components (lower right panel) of Yu2019_QuzDisRh#114, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D107. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_{w}]$-converted eigen-components (upper right panel), and $[P_{w}]$-converted eigen-components (lower right panel) of Yu2019_QzDisRh#102, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D108. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_x]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Yu2019_QzDisRh#104, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D109. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[Ps]$-converted eigen-components (upper right panel), and $[Px]$-converted eigen-components (lower right panel) of Yu2019_QzDisRh#106, which is a quartz dissolution experiment in rhyolite (Yu et al., 2019).
Figure D110. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P]-$converted eigen-components (upper right panel), and $[P]-$converted eigen-components (lower right panel) of Guo&Zhang2016_HB2&4A, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D111. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Guo&Zhang2016_HB3&4A, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D112. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Guo&Zhang2016_HB5&6A, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D113. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[Pw]$-converted eigen-components (upper right panel), and $[Px]$-converted eigen-components (lower right panel) of Guo\&Zhang2016_HB5&7A, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D114. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_{\text{w}}]$-converted eigen-components (upper right panel), and $[P_{\text{x}}]$-converted eigen-components (lower right panel) of Guo&Zhang2016_HB7&8B, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D115. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Guo&Zhang2016_HB9&10A, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D116. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Guo&Zhang2016_HB11&12F, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D117. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_x]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Guo&Zhang2016_HB15&16A, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D118. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_s]$-converted eigen-components (upper right panel), and $[P_s]$-converted eigen-components (lower right panel) of Guo&Zhang2016_HB17&18A, which is a diffusion couple experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D119. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Guo&Zhang2016_An&HB4, which is an anorthite dissolution experiment in haplo-basalt (Guo and Zhang, 2016).
Figure D120. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Guo&Zhang2020_BS1&2C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).

240
Figure D121. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), [Pw]-converted eigen-components (upper right panel), and [Pw]-converted eigen-components (lower right panel) of Guo&Zhang2020_BS3&4C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D122. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P\omega]$-converted eigen-components (upper right panel), and $[P\chi]$-converted eigen-components (lower right panel) of Guo&Zhang2020_BS5&6C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D123. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Guo\&Zhang2020\_BS7\&8C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D124. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Guo&Zhang2020_BS9&10C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D125. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), [Pw]-converted eigen-components (upper right panel), and [Px]-converted eigen-components (lower right panel) of Guo\&Zhang2020_BS11&12C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D126. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Guo&Zhang2020_BS13&14C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D127. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Guo&Zhang2020_BS17&18C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D128. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Guo&Zhang2020_BS19&20C, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D129. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Guo&Zhang2018_BS1&2A, which is a diffusion couple experiment in basalt (Guo and Zhang, 2018).
Figure D130. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_\text{w}]$-converted eigen-components (upper right panel), and $[P_\text{x}]$-converted eigen-components (lower right panel) of Guo&Zhang2018_BS3&4A, which is a diffusion couple experiment in basalt (Guo and Zhang, 2018).
Figure D131. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_x]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Guo&Zhang2018_BS5&6A, which is a diffusion couple experiment in basalt (Guo and Zhang, 2018).
Figure D132. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_X]$-converted eigen-components (upper right panel), and $[P_w]$-converted eigen-components (lower right panel) of Guo & Zhang 2018 BS7 & 8A, which is a diffusion couple experiment in basalt (Guo & Zhang, 2018).
Figure D133. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Guo&Zhang2018_BS9&10A, which is a diffusion couple experiment in basalt (Guo and Zhang, 2018).
Figure D134. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Guo\&Zhang2018_BS11&12A, which is a diffusion couple experiment in basalt (Guo and Zhang, 2018).
Figure D135. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Guo&Zhang2018_BS13&14A, which is a diffusion couple experiment in basalt (Guo and Zhang, 2018).
Figure D136. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P]_x$-converted eigen-components (upper right panel), and $[P]_x$-converted eigen-components (lower right panel) of Guo&Zhang2018_BS17&18A, which is a diffusion couple experiment in basalt (Guo and Zhang, 2018).
Figure D137. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Guo&Zhang2018_BS19&20A, which is a diffusion couple experiment in basalt (Guo and Zhang, 2018).
Figure D138. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_{w}]$-converted eigen-components (upper right panel), and $[P_{X}]$-converted eigen-components (lower right panel) of Guo & Zhang 2020 BS1&2B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D139. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]-\)converted eigen-components (upper right panel), and \([P_X]-\)converted eigen-components (lower right panel) of Guo&Zhang 2020 BS3&4B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D140. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_x]\)-converted eigen-components (lower right panel) of Guo\&Zhang2020_BSS&B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D141. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of Guo & Zhang 2020 BS7&8B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D142. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_0]-\)converted eigen-components (upper right panel), and \([P_3]-\)converted eigen-components (lower right panel) of Guo\&Zhang2020_BS9&10B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D143. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_{w}]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of Guo\&Zhang2020_BS11\&12B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D144. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_x]-\)converted eigen-components (upper right panel), and \([P_x]-\)converted eigen-components (lower right panel) of Guo&Zhang2020_BS13&14B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D145. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_{w}]$-converted eigen-components (upper right panel), and $[P_{X}]$-converted eigen-components (lower right panel) of Guo&Zhang2020_BS17&18B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D146. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_x]\)-converted eigen-components (upper right panel), and \([P_y]\)-converted eigen-components (lower right panel) of Guo&Zhang2020_BS19&20B, which is a diffusion couple experiment in basalt (Guo and Zhang, 2020).
Figure D147. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_a]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of González2017_P050-H0-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - Garcia et al., 2017).
Figure D148. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of González2017_P050-H1-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - García et al., 2017).
Figure D149. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of González2017_P050-H2-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - Garcia et al., 2017).
Figure D150. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]-\)converted eigen-components (upper right panel), and \([P_X]-\)converted eigen-components (lower right panel) of González2017_P100-H0-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - Garcia et al., 2017).
Figure D151. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_x]$-converted eigen-components (lower right panel) of González2017_PI00-H1-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - García et al., 2017).
Figure D152. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_n]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of González2017_P100-H2-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - Garcia et al., 2017).
Figure D153. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_2]-$converted eigen-components (upper right panel), and $[P_X]-$converted eigen-components (lower right panel) of González2017_P300-H0-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - Garcia et al., 2017).
Figure D154. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of González2017_P300-H0-1, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González-Garcia et al., 2017).
Figure D155. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]\)-converted eigen-components (upper right panel), and \([P_X]\)-converted eigen-components (lower right panel) of González2017_P300-H1-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - Garcia et al., 2017).
Figure D156. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of González2017_P300-H2-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González-Garcia et al., 2017).
Figure D157. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of González2017_P300-H2-1, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González-Garcia et al., 2017).
Figure D158. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Gonzalez2017_P300-H2-0, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (Gonzalez - Garcia et al., 2017).
Figure D159. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of González2017_P500-H2-4, which is a diffusion couple experiment between natural shoshonite and a high-K rhyolite (González - García et al., 2017).
Figure D160. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Wang2020_Pyx3, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D161. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Wang2020_Pyx10, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D162. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Wang2020_Pyx11, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D163. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Wang2020_Pyx26, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D164. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Wang2020_Pyx21, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D165. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_a]$-converted eigen-components (upper right panel), and $[P_a]$-converted eigen-components (lower right panel) of Wang2020_Pyx27, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D166. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), \([P_w]-\)converted eigen-components (upper right panel), and \([P_X]-\)converted eigen-components (lower right panel) of Wang2020_Pyx17, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D167. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Wang2020_Pyx20, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D168. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Wang2020_26W01, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D169. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), \([P_{\text{w}}]\)-converted eigen-components (upper right panel), and \([P_{\text{x}}]\)-converted eigen-components (lower right panel) of Wang2020_26W02, which is a lherzolite dissolution experiment in basaltic andesite (Wang et al., 2020).
Figure D170. Concentration profiles of oxide components in wt\% (upper left panel), oxide components in mole\% (lower left panel), $[P_w]$-converted eigen-components (upper right panel), and $[P_X]$-converted eigen-components (lower right panel) of Wang2020_JW03, which is a lherzolite dissolution experiment in ferro-basalt (Wang et al., 2020).
Figure D171. Concentration profiles of oxide components in wt% (upper left panel), oxide components in mole% (lower left panel), $P_w$-converted eigen-components (upper right panel), and $P_X$-converted eigen-components (lower right panel) of Wang2020_JW02, which is a lherzolite dissolution experiment in ferro-basalt (Wang et al., 2020).