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# Trace element partitioning between amphibole and hydrous silicate glasses at 0.6–2.6 GPa

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Abstract Partitioning behavior between amphibole and silicate glass of thirty-three minor and trace elements (Sc, Ti, V, Cr, Co, Rb, Sr, P, Y, Zr, Nb, Cs, Ba, K, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, Ta, Pb, Th, and U) have been determined experimentally. Products of crystallization of hydrous basalt melts from 0.6 GPa/ 860 °C up to 2.6 GPa/970 °C were obtained in a multianvil apparatus. Major and trace element compositions of amphibole and glass were determined with a combination of electron microprobe and laser ablation inductively coupled plasma mass spectrometry. The main mineral phase is calcic amphibole, and the coexisting glass compositions are tonalite, granodiorite, and granite. The compatibility of rare earth elements increase at 915  $\degree$ C and then decrease at 970  $\degree$ C, but the compatibility of most of these elements shows a continued, significant increase with increasing pressure. For high-field strength elements, large ion lithophile elements, actinide compatibility decrease with increasing temperature or pressure, but transition metals show a continued increase in compatibility within the temperature–pressure conditions. From mathematical and graphical fitting, we determined best-fit values for the ideal ionic radius ( $r_0$ , 1.01–1.04 Å), the strain-free partition

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coefficient  $(D_0, 1.18-1.58)$ , and apparent Young's modulus (E, 142–370 GPa) for the M4 site in amphibole according to the lattice strain model. The  $D_0^{MA}$  for rare earth elements rises at 915 °C and then drops at 970 °C at 0.6 GPa. However, the  $D_0^{M4}$  values are positively proportional to the pressure for rare earth elements in the amphibole-glass pairs at 0.6–2.6 GPa and 970 °C. Furthermore, the derived best-fit values for  $r_0^{M4}$  and  $E^{M4}$  are almost constant and trend to increase with rising temperature and pressure, respectively. The partition coefficient is distinctly different for different melt compositions. The rare earth elements become more enriched in amphibole if the quenched glass is granodiorite or granite compared to the tonalitic glasses.

Keywords Amphibole - Silicate glass - Trace elements - Partition coefficients · Lattice strain model

## 1 Introduction

The partition coefficient describes the distribution of an element between a mineral and melt. The Nernst partition coefficient is defined as  $D_i$ = (concentration of element *i* in mineral)/(concentration of element  $i$  in melt). One could obtain quantitative understandings on igneous evolution by estimating the parental melt compositions with relevant D values between minerals and melts (Lundstrom et al. [2000](#page-14-0); Landwehr et al. [2001](#page-14-0); Frei et al. [2009](#page-14-0); Severs et al. [2009](#page-14-0); Parker et al. [2010;](#page-14-0) Chang et al. [2016;](#page-13-0) Cui et al. [2017](#page-13-0)). However, due to the effects of temperature, pressure and composition, mineral/melt D values show enormous variations (Blundy and Wood [2003;](#page-13-0) Adam and Green [2006](#page-13-0); Severs et al. [2009;](#page-14-0) Sun [2018\)](#page-14-0).

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<span id="page-1-0"></span>Most of the partition coefficient studies focus on minerals and silicate melts. These minerals include amphibole (Adam and Green [2006](#page-13-0); Tiepolo et al. [2007](#page-15-0); Nandedkar et al. [2016](#page-14-0); Shimizu et al. [2017](#page-14-0)), clinopyroxene (Adam and Green [2006;](#page-13-0) Hill et al. [2011](#page-14-0); Sun and Liang [2012](#page-14-0); Mollo et al. [2017\)](#page-14-0), orthopyroxene (Adam and Green [2006](#page-13-0); Frei et al. [2009;](#page-14-0) Parker et al. [2010](#page-14-0); Sun and Liang [2013a](#page-14-0)), garnet (Adam and Green [2006;](#page-13-0) Draper and Westrenen [2007;](#page-14-0) Sun and Liang [2013b\)](#page-14-0), plagioclase (Severs et al. [2009;](#page-14-0) Sun et al. [2017](#page-14-0)), olivine (Adam and Green [2006](#page-13-0); Sun and Liang [2013b;](#page-14-0) Stead et al. [2017](#page-14-0)), and some infrequent crystals such as rutile (Klemme et al. [2005](#page-14-0); Xiong et al. [2005\)](#page-15-0), and apatite (Prowatke and Klemme [2006](#page-14-0)). Additionally, few published literatures showed the distribution of trace elements between amphibole, clinopyroxene, garnet, apatite and carbonatite melts (Sweeney et al. [1992,](#page-14-0) [1995;](#page-14-0) Blundy and Dalton [2000;](#page-13-0) Klemme and Dalpe´ [2003;](#page-14-0) Dasgupta et al. [2009\)](#page-13-0).

Different techniques have been used to determine partition coefficient of trace elements between minerals and melts. Several studies have measured trace elements in minerals and surrounding glassy matrix. The first kind of studies measured the contents of trace elements in minerals and the surrounding glassy matrix (Arzamastsev et al. [2009;](#page-13-0) Severs et al. [2009;](#page-14-0) Olin and Wolff [2010](#page-14-0); Mollo et al. [2017\)](#page-14-0). Moreover, the melt inclusion-mineral (MIM) technique was introduced to determine partitioning behavior of elements (Thomas et al. [2002](#page-14-0); Zajacz and Halter [2007](#page-15-0); Severs et al. [2009;](#page-14-0) Nardi et al. [2013\)](#page-14-0). Finally, minerals and melts from experimental products were used to determine the partitioning of trace elements (Blundy and Dalton [2000;](#page-13-0) Dalpe´ and Baker [2000;](#page-13-0) Adam and Green [2006](#page-13-0); Frei et al. [2009](#page-14-0); Parker et al. [2010](#page-14-0); Hill et al. [2011\)](#page-14-0). These methods have been used to determine partition coefficients

**Table** 

for the trace elements in various compositions of minerals and co-existing melts. Generally, mineral phases-melt systems with many minerals were investigated rather than a single mineral-melt system in most of the previous studies (Adam and Green [2006](#page-13-0); Arzamastsev et al. [2009](#page-13-0); Severs et al. [2009;](#page-14-0) Wan et al. [2009](#page-15-0)). For example, Adam and Green ([2006\)](#page-13-0) discussed the partition coefficients of trace elements between amphibole, clinopyroxene, orthopyroxene, garnet, olivine and coexisting melt in the same system. Furthermore, it is well known that partition coefficient depends on the chemical compositions of mineral and melt, on pressure and temperature and on  $H_2O$ -content of the melt (Blundy and Wood [2003;](#page-13-0) Adam and Green [2006](#page-13-0); Severs et al. [2009](#page-14-0); Parker et al. [2010](#page-14-0); Hill et al. [2011](#page-14-0); Sun et al. [2017\)](#page-14-0).

In addition, a theoretical framework based on the lattice strain model (Brice [1975](#page-13-0)) was provided for trace element partitioning (Blundy and Wood [1994\)](#page-13-0). So far, the theory has been applied successfully to trace element partitioning between clinopyroxene (Wood and Blundy [1997](#page-15-0); Adam and Green [2006;](#page-13-0) Sun and Liang [2012\)](#page-14-0), garnet (Draper and Westrenen [2007;](#page-14-0) Westrenen and Draper [2007;](#page-15-0) Sun and Liang [2013b\)](#page-14-0), and amphibole (Adam and Green [2006](#page-13-0); Tiepolo et al. [2007](#page-15-0); Wan et al. [2009](#page-15-0)) and the coexisting melt with each other. These studies have shown that partition coefficients vary as a function of crystal and melt composition, pressure and temperature. The influence of melt composition and temperature on the distribution of trace elements has been studied abundantly (Klein et al. [1997](#page-14-0); Tiepolo et al. [2000](#page-14-0); Nandedkar et al. [2016](#page-14-0); Shimizu et al. [2017;](#page-14-0) Sun [2018](#page-14-0)). However, the systematic study of the effect of pressure on the distribution of trace elements is relatively lacking.



Sample number 10084 from Zhou et al. ([1998\)](#page-15-0)

Major- and trace-elements concentration are given in wt% and ppm, respectively

<span id="page-2-0"></span>

Table 2 Run conditions and phase compositions of amphibole and glass in experimental products Table 2 Run conditions and phase compositions of amphibole and glass in experimental products

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Amp amphibole, Apa apatite, Sp spinel, Ab albite, Gl quenched glass

Amp amphibole, Apa apatite, Sp spinel, Ab albite, Gl quenched glass

<span id="page-3-0"></span>Table 3 Trace element data of experimental products for hydrous basalt

RUN No.	10084-1		10084-2		10084-3		10084-4	
Phase $(n)$	Amp $(4)$	Gl(4)	Amp $(5)$	Gl(6)	Amp $(7)$	Gl(4)	Amp $(7)$	Gl(5)
Sc	20.6(0.9)	18.5(0.2)	20.5(1.5)	18.2(2.9)	29.3 (4.9)	15.8(5.8)	21.4(1.6)	3.29(1.21)
Ti	9441 (901)	8750 (485)	13462 (1093)	11,439 (1076)	20,913 (2200)	11,500 (401)	21,733 (1791)	5144 (403)
V	133(10)	126(6)	159(15)	136(35)	351 (57)	155(62)	298 (23)	9.92(0.87)
Cr	131 (26)	125(9)	168(17)	115(39)	353 (50)	194 (59)	293 (29)	24(13)
Rb	33.4 (2.3)	31.4(2.3)	65.9(5.0)	77.6(6.1)	24.3(1.9)	68.9 (5.8)	20.1(1.8)	183.6(7.6)
Sr	929 (80)	686 (48)	783 (38)	789 (76)	850 (94)	1072 (129)	761 (57)	1390 (51)
Y	15.9(1.2)	11.4(0.6)	47.7(4.1)	23.3(2.5)	23.6(1.9)	21.6(2.1)	21.4(1.0)	14.7(1.4)
Zr	162(19)	139(12)	229 (18)	252 (20)	187(15)	284 (26)	142(10)	356 (12)
Nb	36.4(2.7)	33.9(2.7)	53.2 $(2.8)$	48.7 (2.5)	56.4 (3.4)	69.4 $(4.3)$	49.3 $(4.1)$	84.0 (4.6)
<b>Cs</b>	3.12(0.65)	2.09(0.07)	5.57(0.69)	6.67(0.55)	1.96(0.19)	5.43 (1.66)	1.46(1.14)	15.3(1.0)
Ba	479 (48)	409 (44)	586 (41)	600(57)	416 (399)	780 (62)	389 (31)	1123 (34)
K	10,612 (1204)	9201 (297)	17,074 (1105)	18,317 (2404)	7885 (656)	15,386 (1333)	10,713 (1581)	33,763 (715)
La	15.7(1.5)	68.2(0.7)	42.5 $(2.5)$	69.2 (6.3)	27.2(2.3)	57.4 (3.2)	24.6(1.6)	76.3 (8.2)
Ce	45.6(6.2)	121(1)	84.8 (4.5)	114(11)	59.8 (5.1)	102(8)	57.7 (6.8)	127(10)
Pr	7.88(0.71)	12.2(0.1)	9.39(1.43)	11.3(1.2)	7.92(0.18)	11.3(1.8)	7.44(1.03)	12.1(1.3)
Nd	39.1(2.4)	45.4(0.5)	40.7(3.5)	38.7(2.2)	35.1(2.8)	42.2(2.5)	34.9 (1.2)	39.8 (2.8)
Sm	4.60(0.87)	4.10(0.28)	8.76 (1.98)	6.61(1.28)	8.56 (1.57)	7.59 (1.90)	7.25(1.18)	7.55(0.24)
Eu	1.70(0.37)	1.45(0.31)	2.20(0.40)	1.59(0.33)	2.78(0.71)	2.20(0.97)	2.50(0.64)	2.79(0.89)
Gd	5.22 (0.46)	4.78 (0.09)	6.78(1.51)	4.51(0.97)	7.97(0.01)	6.57(0.97)	7.40(0.14)	5.07 (0.92)
Tb	0.55(0.02)	0.43(0.01)	0.90(0.23)	0.59(0.13)	1.15(0.46)	1.05(0.38)	1.09(0.40)	0.98(0.38)
Dy	3.13(0.08)	2.55(0.28)	4.93(0.82)	3.66(0.65)	5.68 (0.46)	4.63(0.74)	5.03(0.22)	3.83(0.37)
Ho	1.18(0.09)	0.79(0.02)	0.81(0.15)	0.41(0.10)	0.98(0.27)	0.97(0.28)	1.12(0.28)	0.83(0.35)
Er	1.14(0.13)	0.82(0.16)	1.96(0.31)	0.97(0.34)	2.18(0.71)	2.30(0.69)	1.97(0.44)	1.87(0.46)
Tm	0.14(0.02)	0.11(0.07)	0.30(0.04)	0.19(0.03)	0.37(0.15)	0.36(0.13)	0.39(0.11)	0.57(0.24)
Yb	2.45(0.20)	1.81(0.21)	1.49(0.36)	1.40(0.36)	1.97(0.42)	1.85(0.84)	2.30(1.04)	2.17(0.75)
Lu	0.12(0.01)	0.09(0.01)	0.26(0.04)	0.33(0.04)	0.27(0.12)	0.28(0.04)	0.36(0.13)	0.54(0.26)
Hf	3.32 (0.66)	2.93(0.02)	5.70 (0.89)	5.26(0.05)	5.32(0.73)	6.32(0.99)	4.49 (0.04)	7.14(1.26)
Ta	2.30(0.56)	1.98(0.18)	3.24(0.23)	3.26(0.48)	3.01(0.41)	4.98 (0.86)	2.93(0.78)	6.24(0.67)
Pb	14.3(1.6)	11.9(0.4)	24.1(2.0)	29.1(2.8)	9.04(0.76)	23.2(2.8)	8.87 (1.43)	99.3(6.1)
Th	3.82(0.61)	3.08(0.22)	9.26(0.61)	9.24(1.09)	2.58(0.62)	7.01(0.76)	1.71(0.19)	12.9(1.7)
U	1.42(0.26)	1.29(0.18)	2.74(0.56)	2.82(0.74)	1.08(0.48)	2.49 (0.77)	0.82(0.60)	5.22(0.58)
RUN No.	10084-5			10084-6		10084-7		
$\mathbf n$	Amp $(5)$	Gl(5)		Amp $(5)$	Gl(9)	Amp $(4)$		Gl(5)
Sc	25.7 (2.9)		4.32(0.22)	21.6(0.9)	4.51(0.68)		19.1(2.3)	3.78(0.76)
Ti	20,776 (1390)		3954 (257)	16,039 (1896)	4540 (413)		34,230 (10,395)	12,456 (1255)
V	260(24)		7.79(1.06)	232 (20)	4.97(0.84)		150(10)	4.82(0.30)
Cr	227 (23)		7.21 (1.07)	214(15)	6.75(0.39)		121(15)	12.2(1.4)
Rb	17.5(1.2)		175(7)	18.9(1.5)	208(7)		14.6(1.6)	173(3)
Sr	731 (68)		1495 (41)	731 (53)	1745 (66)		909 (46)	2154 (55)
Y	20.2(2.0)		13.1(1.6)	21.8(1.3)	14.6 $(1.4)$		19.4(1.9)	
Zr	148 (12)		427(10)	150 (17)	471 (16)		132 (12)	567 (37)
Nb	45.6(2.8)		95.1 (2.5)	40.0(4.3)	123(6)		34.3 (2.8)	136(9)
Cs	1.38(0.90)		16.7(0.3)	0.95(0.56)	17.2(0.9)		3.05(0.47)	17.3(1.1)
Ba	383 (26)		1369 (23)	418 (32)	1549 (58)		503 (25)	994 (191)
K	8962 (817)		34,957 (846)	9936 (1725)	32,476 (1521)		13,245 (1110)	34,304 (1518)
La	17.9(1.4)		66.3 (3.9)	16.4(2.1)	84.2 (4.0)		12.1(1.3)	80.3 (4.2)





Trace element concentrations are reported in ppm  $(\mu g/g)$ . *n* is the number of analyses. The numbers in the parentheses are standard deviation. For example, 20,776 (1390), 25.7 (2.9), and 4.32 (0.22) should be read as 20,776  $\pm$  1390, 25.7  $\pm$  2.9, and 4.32  $\pm$  0.22, respectively

This study investigated the partitioning of trace elements between the crystalline amphibole and silicate liquid from the experimental products of crystallization of hydrous basalt melts at 0.6–2.6 GPa and 860–970 °C. A combination of electron microprobe (EMP) and laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) was used to analyze major and trace elements in experimentally produced amphiboles and quenched glasses. The abundances of rare earth elements (REE), highfield strength elements (HFSE), large ion lithophile elements (LILE), actinides, and the transition metals in experimental phases were determined and the data were used to calculate the partition coefficients between amphiboles and glasses. In addition, the ideal radius, the maximum partition coefficient and the apparent Young's modulus of the M4 site of amphibole were estimated assuming that all  $REE^{3+}$  occupy this site.

## 2 Experimental strategy and analytical techniques

## 2.1 Starting material and experimental procedures

The starting material used in the crystallization experiments is a natural hydrous basalt (10,084) with modal mineralogy plagioclase (40%), iddingsitized olivine (25%),

clinopyroxene (20%), matrix (10%) and minor iron titanium oxide (5%) from the Yichuan–Ruyang region in west Henan province, China. Table [1](#page-1-0) shows the composition of the starting material (Zhou et al. [1998\)](#page-15-0).

Two series of experiments (temperature series and pressure series) were conducted in the multi-anvil apparatus in the Key Laboratory for High Temperature and High Pressure Study of the Earth's Interior, Institute of Geochemistry, Chinese Academy of Sciences. Run conditions and the experimental protocol and configuration of charges for multi-anvil experiments were same with those described by Zhang et al. [\(2019\)](#page-15-0). Fine-ground powder samples were loaded into 3 mm long graphite containers with inner diameter 4 mm, and placed between 1 mm thick graphite discs at each end. The pressure transmitting medium was pyrophyllite. In the ''temperature series'', charges were held at 0.6 GPa and 1460  $\degree$ C for 1 h, then rapidly cooled isobarically to crystallization temperatures of 860 $\degree$ C, 915 °C, or 970 °C, and held for 100 h before quenching. In the ''pressure series'', the charges were brought to superliquidus conditions (1460 °C and 0.6 GPa, 1490 °C and 1.1 GPa, 1515 °C and 1.6 GPa, 1540 °C and 2.1 GPa, or 1570 °C and 2.6 GPa) for 1 h, and then rapidly and isobarically cooled to an identical crystallization temperature of 970  $\degree$ C and annealed for 100 h. All samples were quenched by shutting off the electric power. Experimental

 $100~\mu{\rm m}$ 



<span id="page-6-0"></span> $\blacktriangleleft$  Fig. 1 BSE images of experimental charges at 0.6–2.6 GPa and 860–970 °C illustrating coexisting minerals and glass and the increase in crystal size of amphibole with increasing pressure or temperature. Amp amphibole, Apa apatite, Gl glass

conditions are reported in Table [2.](#page-2-0) Run products were mounted in an epoxy disc and carefully polished for EMP and LA-ICP-MS analysis.

#### 2.2 Analytical techniques

Major element compositions of phases were analyzed with the JEOL JXA-8100 electron microprobe at the Institute of Geology and Geophysics, Chinese Academy of Sciences. An accelerating voltage of 15 kV and beam current of 20 nA were employed. For mineral analysis, the beam diameter was  $1-5$  µm and a slightly defocused beam of 10 µm diameter was used for glass. Na and K were counted in the first pass to reduce the problem of Na and K loss during analyses (Morgan and London [2005](#page-14-0)). Major element compositions of amphibole and glass in the experimental products are reported in Table [2](#page-2-0). Experimental charges were analyzed for trace elements by LA-ICP-MS (Agilent 7700X) at the State Key Laboratory of Ore Deposit Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences. Laser sampling was performed using a Coherent GeolasPro laser ablation system (193 nm wavelength). Analytical methods are available in Huang et al. [\(2016](#page-14-0)). Each analysis incorporates an approximately 15 s background acquisition (gas blank) followed by 50 s data acquisition from the sample. Analytical spots  $(44 \mu m)$  were

ablated by 160 successive laser pulses (4 Hz). Every 5–6 sample analyses were followed by one analysis of NIST SRM 610 in order to correct the time-dependent drift of sensitivity and mass discrimination. Data reduction was performed by ICPMSDataCal (Liu et al. [2008\)](#page-14-0). Trace element compositions of amphibole and glass in the experimental products are summarized in Table [3.](#page-3-0)

## 3 Results

#### 3.1 Description of run products

All of seven runs yielded amphibole-glass pairs that were suitable for the determination of trace element partition coefficients (Fig. 1, Table [2](#page-2-0)). The experimental phase assemblages are dominated by amphibole (Amp), glass (Gl). Some experiments also include accessory apatite (Apa), spinel (Sp), albite (Ab) or magnetite (Mt). Backscattered electron (BSE) images of the experimental charges illustrate the increasing crystal size of euhedral amphibole with increasing pressure or temperature. At 0.6 GPa and 860  $\degree$ C, amphibole is about 80 µm in the maximum size and distributed within the glass (Fig. 1a). Amphibole-glass contacts are straight without any resorption textures. Furthermore, at 0.6 GPa and 970 °C, euhedral amphibole crystal of  $\sim 100 \mu m$  formed together with slightly smaller apatite (Fig. 1c). Amphibole crystal about 300 µm formed, which coexists with large, block apatite crystals at 2.6 GPa and 970 °C (Fig. 1g).



Fig. 2 Classifications of amphibole and glass in the experimental products. a Calcium amphibole and their compositional boundaries, showing the crystalline amphiboles at 0.6–1.1 GPa and 1.6–2.6 GPa fall in pargasite and sadanagaite fields, respectively. The calcium amphiboles defined by <sup>B</sup>(Ca +  $\Sigma M^{2+}$ )/ $\Sigma B \ge 0.75$  and <sup>B</sup>Ca/ $\Sigma B \ge \Sigma M^{2+}$ / $\Sigma B$ . b CIPW normative An-Ab-Or plot for quenched glasses, showing the run liquids fall in tonalite and granodiorite fields at 0.6 GPa but fall in granodiorite to granite with increasing pressure. 0.6/860 means crystallization pressure P (GPa) and temperature  $T$  (°C) in the legend

Table 4 Amphibole-glass partition coefficients

#### <span id="page-7-0"></span>3.2 Amphibole and glass compositions

Amphibole and glass have homogeneous major and trace element compositions in each run (Table [2,](#page-2-0) [3](#page-3-0)), and their classification is shown in Fig. [2.](#page-6-0) According to the recommendations of Hawthorne et al. ([2012\)](#page-14-0), all the experimental amphiboles belong to the calcic amphibole category and are classified pargasite/sadanagaite and have  $Mg# =$ 50–60 [Mg# = molar (100  $\times$  MgO/(MgO + FeO))]. Figure [2](#page-6-0)a shows the crystalline amphiboles at 0.6–1.1 GPa and 1.6–2.6 GPa fall in pargasite and sadanagaite fields, respectively. From the CIPW normative An-Ab-Or plot for quenched glasses, Fig. [2](#page-6-0)b shows the run liquids fall in tonalite and granodiorite fields at the lower pressure (0.6 **Fig. 3** Trace elements partition coefficients  $(D_i)$  between amphiboles and glasses from this study and available literature studies. a, b Partition coefficients for REE; c, d Partition coefficients of high-field strength elements (Nb, Ta, Zr, Hf, Ti); e, f Partition coefficients of large ion lithophile elements K, Rb, Cs, Sr and Ba; g, h Partition coefficients for actinides (U, Th) and Pb; i, j Partition coefficients of transition metals (Sc, V, Cr). The legends of temperature series runs and the pressure series runs are shown in figure a and b, respectively. 0.6/860 means crystallization pressure  $P$  (GPa) and temperature  $T$  (°C) in the legend. *Open symbols* data from previous literature. 1.0/ 850, 900: Klein et al. ([1997\)](#page-14-0); 0.7/860, 890, 920, 950: Nandedkar et al. ([2016\)](#page-14-0); 1.25/900: Qian and Hermann [\(2013](#page-14-0)); 1.5/950: Wan et al. ([2009\)](#page-15-0)



Uncertainties are the numbers in the parentheses and are reported as standard deviation with respect to the average and are calculated by propagating uncertainty of the individual phase.  $T_{im}$ , and  $K_m$  indicate partition coefficients based on microprobe measurements



<span id="page-9-0"></span>

Fig. 3 continued

GPa) but in granodiorite to granite at the higher pressures (1.1–2.6 GPa). Most trace elements are present in smaller amounts in amphibole than those in coexisting glass, but the concentrations of medium to heavy rare earth elements of amphibole are greater than those in the glass (Table [3](#page-3-0)).

## 4 Discussion

#### 4.1 Attainment of equilibrium

The attainment or close approach to thermodynamic equilibrium of crystallization during the experiment is an important prerequisite for accurate trace element partitioning coefficient. Although we did not perform reversal experiments, there are two arguments that suggest close approximation to equilibrium. Firstly, the experimental duration (100 h) of our study is longer than previous studies that have observed attainment of equilibrium (Klein et al. [1997;](#page-14-0) Klemme et al. [2002](#page-14-0); Adam and Green [2006](#page-13-0); Nandedkar et al. [2014\)](#page-14-0). Secondly, the homogeneous composition of the quenched experimental products as indicated by the average standard deviations of less than  $< 5\%$ for major elements (Table [2\)](#page-2-0) and of less than  $\lt 10\%$  for trace elements (Table [3\)](#page-3-0) further support our interpretation that major and trace element equilibrium was achieved.

## 4.2 Trace element partitioning

Amphibole-glass partition coefficients  $(D_i)$  calculated from the average trace element concentrations are listed in Table [4](#page-7-0) and summarized in Fig. [3.](#page-7-0) Uncertainties in the coefficients are estimated by error propagation from analytical uncertainties in the individual phase.



4.2.1 Rare earth elements

The amphibole–glass partition coefficients for REE in the temperature series products are illustrated in Fig. [3a](#page-7-0). The light rare earth elements (LREE: La–Nd) are incompatible elements with the exception of Nd at 0.6 GPa and 915  $^{\circ}$ C. In contrast, medium to heavy rare earth elements (MREE-HREE: Sm–Lu) are compatible apart from Lu at 0.6 GPa and 915 °C, and Er and Lu at 0.6 GPa and 970 °C. This observation is consistent with the results from Nandedkar et al. [\(2016](#page-14-0)). The compatibility of HREE is slightly increasing with increasing atomic number except the Yb at 0.6 GPa and 915 °C. The behavior of Y is analogous to that of HREE due to its identical valence and similar cation radius. It is worthy note that the  $D_{\text{REE}}$  (with the exception of Yb and Lu) slightly increase at 915  $\degree$ C and then decrease at 970 $\degree$ C.

Moreover, most REE show a continuous, smooth change in their  $D$  values for amphibole-glass with increasing  $P$  in the pressure series products (Fig. [3](#page-7-0)b). All of the LREE are incompatible elements, and the  $D<sub>LREE</sub>$  decrease with increasing pressure at isothermal 970 °C. But almost all of MREE and HREE are compatible elements apart from Sm at 1.1 GPa, Eu, Tm and Lu at 1.1 GPa to 2.1 GPa. Additionally, the most compatible rare earth element in the amphibole structure is dysprosium (Dy) in agreement with previous studies (Klein et al. [1997](#page-14-0); Nandedkar et al. [2016](#page-14-0)).

Fig. 4 Onuma diagrams, log  $(D)$  versus cation radius, for REE partition coefficients between amphibole and glass. Curves are fits of Eq. [1](#page-12-0) to the data. Measuring these partition coefficients for REE show parabolic behavior. The  $\overline{REE}^{3+}$  cations enter M4 site in amphibole from the temperature series products (a–c) and the pressure series products (d–g)



<span id="page-11-0"></span>The partition coefficients for some MREE (Eu, Tb, and Dy) and HREE (Tm and Yb) are positively proportional to pressure at  $0.6-2.6$  GPa and 970 °C.

## 4.2.2 High-field strength elements

HFSE are tetra- or penta-valent cations such as Nb, Ta, Zr, Hf, and Ti. The amphibole–glass partition coefficients for HFSE in the temperature series products are plotted in Fig. [3](#page-7-0)c. The partition coefficients of HFSE are  $> 1$  at 0.6 GPa and 860  $^{\circ}$ C. But the HFSE become weakly compatible with incompatible as the temperature increases from 860 to 970  $\degree$ C at 0.6 GPa, with the exception of Ti, which is compatible in the amphibole, as also shown in previous studies (Tiepolo et al. [2007](#page-15-0); Nandedkar et al. [2016\)](#page-14-0).

Nb, Ta, Zr, and Hf are incompatible and their partition coefficients are generally  $\lt 1$  in the pressure series, but the  $D_{\text{Ti}}$  is greater than one (Fig. [3d](#page-7-0)). In addition, the HFSE (with the exception of Ti) partition coefficients tend to decrease as the pressure increases from 0.6 to 2.6 GPa at 970  $^{\circ}$ C. In summary, the partition coefficients for HFSE increase with decreasing temperature or pressure, and the Ti is more compatible than other HFSE.

#### 4.2.3 Large ion lithophile elements

Besides the result of 0.6 GPa and 860  $^{\circ}$ C, the LILE are incompatible elements with partition coefficients generally below 1 in the temperature series and the pressure series (Fig. [3e](#page-7-0), f). In the temperature series products, the LILE are weakly compatible with moderately incompatible with increasing temperature at 0.6 GPa. The partition coefficient of the potassium (K) in this study indicates the same trend relative to Klein et al. [\(1997](#page-14-0)) and decreases with increasing temperature, but Nandedkar et al. ([2016\)](#page-14-0) show an opposite trend. In the pressure series products, the partition coefficients for most LILE exhibit a negative correlation with pressure at 0.6–2.6 GPa and 970 °C. Moreover,  $D_{\text{LIE}}$  from the pressure series experiments show a uniform pattern when plotted against the atomic number in agreement with previous studies (Wan et al. [2009;](#page-15-0) Nandedkar et al. [2016](#page-14-0)), with the exception of Cs, Sr, and Ba in the run of 2.6 GPa and 970  $\degree$ C and Cs, Sr in the study of Oian and Hermann [\(2013](#page-14-0)). Another similarity is the maximum partition coefficient for Sr except for Ba at 2.6 GPa and 970 °C.

## 4.2.4 Actinides

Uranium (U) and thorium (Th) amphibole–glass partition coefficients are reported in Fig. [3](#page-7-0)g, h together with the partition coefficient for Pb. Besides the result of 0.6 GPa and  $860^{\circ}$ C, the actinides elements are incompatible

**Table 5** Fitting parameters used in the lattice strain model for  $REE^{3+}$  cations in amphibole

P/GPa	$T$ /°C	$r_0/A$	$\sigma$	$D_0$	$\sigma$	E/GPa	$\sigma$	Melt composition	References
0.6	860	1.01	0.01	1.38	0.1	142	31	Tonalite	This study
0.6	915	1.04	0.03	1.52	0.1	162	26	Granodiorite	
0.6	970	1.04	0.01	1.18	0.08	165	41	Tonalite	
1.1	970	1.04	0.02	1.26	0.1	273	56	Granodiorite	
1.6	970	1.03	0.03	1.41	0.09	271	22	Granite	
2.1	970	1.03	0.04	1.44	0.1	370	69	Granite	
2.6	970	1.01	0.01	1.52	0.1	238	25	Granite	
1.0	900	1.03	0.007	1.7	0.8	110	80	Andesite	Green and Pearson (1985)
1.5	1000	0.99	0.03	1.3	0.2	204	81	Andesite	Brenan et al. $(1995)$
1.5	1150	1.03		0.6		390	-	Basanite	LaTourrette et al. (1995)
1.0	800	1.03	0.01	2.66	0.19	170	60	Tonalite	Klein et al. (1997)
1.0	850	1.03	0.01	2.29	0.1	250	60		
1.0	900	1.03	0.01	1.75	0.1	300	80		
1.0	1025	1.035	-	0.64	-	300	-	Basanite	Adam and Green (2006)
2.0	1050	1.03	-	0.41	-	320	-		
1.5	950	0.995	0.006	1.32	0.03	233	35	Granodiorite	Wan et al. (2009)
0.7	950	1.02	0.001	2.63	0.03	339	9	Andesite	Nandedkar et al. (2016)

 $\sigma$  indicates 1 sigma standard deviation of these fitting parameters. (-) no value available

<span id="page-12-0"></span>

Fig. 5 Change of fitting parameters  $r_0$ ,  $D_0$  of the REE with increasing T at isobaric 0.6 GPa (a), and increasing P at isothermal 970 °C (b)

elements with partition coefficients below 1 in the temperature series and the pressure series. Th and U show decreasing partition coefficients with increasing temperature with  $D_{\text{Th}}$  varying from 1.24 to 0.37 and  $D_{\text{U}}$  from 1.1 to 0.43. Similar to temperature series, U and Th show a consistent decrease in partition coefficients between 0.6 and 2.6 GPa.  $D_{\text{Pb}}$  displays a decrease with increasing temperature or pressure. However, our data show substantially higher partition coefficients for actinides than that in previous studies (Klein et al. [1997](#page-14-0); Nandedkar et al. [2016\)](#page-14-0). Probably, the result has to do with the relatively lower silica concentration of glasses from this experiment.

## 4.2.5 Transition metals

The partition coefficients for transition metals are rarely reported in the literature. Three transition metals, Sc, V and Cr, are investigated in this study (Fig.  $3i$  $3i$ , j); all of them are compatible elements and their partition coefficients positively correlate with temperature, with  $D_{\text{Sc}}$ ,  $D_{\text{V}}$ , and  $D_{\text{Cr}}$ ranging from 1.11 to 1.85, 1.06 to 2.26, and 1.05 to 1.82 at 860–970  $\degree$ C, respectively. Similarly, their partition coefficients trend to increase with increasing pressure. For V and Cr, there is a notable increase in partition coefficients between 0.6 and 1.1 GPa. Nandedkar et al. [\(2016](#page-14-0)) suggested the substantial increase of D for transition metals is related to change of the melt structure.

## 4.3  $D_{\text{REE}}^{\text{Amp-Gl}}$  with the lattice strain model

The influence of cation radius (Shannon [1976](#page-14-0)) on partitioning behavior for trace elements has been described by many publications (Onuma et al. [1968;](#page-14-0) Brice [1975](#page-13-0); Blundy and Wood [1994](#page-13-0); Wood and Blundy [1997](#page-15-0); Blundy and Wood [2003](#page-13-0)). The relationship, Lattice Strain Model-LSM,

$$
D_{i} = D_{0} \exp \frac{-4\pi EN_{A} \left[\frac{r_{0}}{2} (r_{i} - r_{0})^{2} + \frac{1}{3} (r_{i} - r_{0})^{3}\right]}{RT}
$$
(1)

relates the Nernst partition coefficients of elements  $i(D_i)$ for isovalent cations in crystals and melts as a function of cation radius  $(r_i)$ , the strain-free partition coefficient  $(D_0)$ that has an ionic radius  $r_0$  equal to the strain-free lattice size of the crystallographic site, and an elastic constant  $E$  (apparent Young's modulus) that is specific to the host cation site.  $N_A$  is Avogadro's constant, R is the universal gas constant, and  $T$  is temperature in Kelvin.

According to the lattice strain model, the  $r_0$ ,  $D_0$ , and  $E$  are estimated by combining with the partition coefficients of trivalent REE between amphiboles and silicate glasses. Partition coefficients versus cation radius for the  $REE<sup>3+</sup>$  cations entering M4 site of amphibole are plotted in Fig. [4](#page-9-0), these Onuma diagrams display the parabolic shape. A set of parabolas have the following ranges in fitting parameter values for the M4 site of amphibole,  $r_0$ 1.01–1.04 A,  $D_0$  1.18–1.38, and E 142–165 GPa at 0.6 GPa and  $860-970$  °C (Fig. [4a](#page-9-0)–c, Table [5\)](#page-11-0). Although some previous studies suggested that the  $D_0$  decreases with increasing temperature (Klein et al. [1997](#page-14-0); Nandedkar et al. [2016](#page-14-0); Shimizu et al. [2017](#page-14-0)), our  $D_0$  has the maximum value at 0.6 GPa and 915 °C rather than at 860 °C (Fig. 5a). Nevertheless, the  $D_0$  trends to decrease at the range of 860–970 °C. Furthermore, Shimizu et al. ([2017\)](#page-14-0) verified that  $D_0$  positively correlates with the silica content of the coexisting glass. The maximum REE partition coefficient  $D_0$  corresponds with the highest silica content of the coexisting glass at 0.6 GPa and 915 °C. For example, the REE are easier enriched in amphibole if the quenched glass is granodiorite or granite in this study, and the  $D_0$  values are similar to the result of Wan et al. [\(2009](#page-15-0)). However, the  $D_0$  between amphibole and tonalitic glass (Klein et al. [1997](#page-14-0)) and between amphibole and andesitic glass (Green

<span id="page-13-0"></span>and Pearson, [1985;](#page-14-0) Brenan et al. 1995; Nandedkar et al. [2016\)](#page-14-0) are greater compared to amphibole-basaltic glass pair (LaTourrette et al. [1995](#page-14-0); Adam and Green 2006). Thus, glass composition exerts the dominant control on the incorporation of REE on the M4 site of amphibole at 0.6 GPa and  $860-970$  °C.

In addition, the fitting parameters are  $r_0$  1.01–1.04 Å,  $D_0$ 1.18–1.52, and E 165–370 GPa at 0.6–2.6 GPa and 970 °C (Fig. [4](#page-9-0)c–g, Table [5](#page-11-0)). Amphibole-glass REE partition coefficients have been shown to negatively correlate with P in some previous studies (Adam and Green 1994, 2003; Dalpé and Baker  $2000$ , but our  $D_0$  increases with increasing pressure (Fig. [5](#page-12-0)b). In the 'melt model' from Shimizu et al. [\(2017](#page-14-0)), the  $D_0$  negatively correlates with Ti and Ca contents in the glass, and the pressure series shows the same result in this study. Thus, the effect of pressure is significant for the partition coefficient of REE between amphibole and coexisting glass at  $0.6-2.6$  GPa and 970 °C. The ideal radius  $r_0$  of amphibole M4 is almost constant both temperature series and pressure series, and the E trends to increase with increasing temperature or pressure (with the exception of 2.6 GPa).

## 5 Conclusions

The calcic amphibole is a sufficiently dominant mineral phase from crystallization of hydrous basalt melts at 0.6–2.6 GPa and 860–970 °C, and the coexisting glass compositions are tonalite, granodiorite, and granite.

The LREE are dominantly distributed in the glass rather than the amphibole in this study. In contrast, most MREE and HREE are compatible in amphibole. These patterns are in good agreement with previous findings, with the REE becoming increasingly compatible with decreasing ionic radius (Klein et al. [1997](#page-14-0); Wan et al. [2009;](#page-15-0) Nandedkar et al. [2016\)](#page-14-0). It is notable that at 0.6 GPa, the  $D_{\text{REE}}$  (with the exception of Yb and Lu) slightly increase at 915 °C but then decrease at 970 °C. Similarly, the  $D_0^{M4}$  for REE increases at 915 °C and then decreases at 970 °C. However, the  $D_0^{M4}$  values positively correlate with the pressure for REE in the amphibole-glass pairs. Furthermore, the derived best-fit values for  $r_0^{M4}$  and  $E^{M4}$  are almost constant and trend to increase with rising temperature or pressure, respectively. Although the LILE, HFSE (except the Ti), and actinides show a continuous decrease in compatibility with increasing temperature or pressure, the D of transition metals is inversely proportional to the T-P conditions in these experimental series. The experimental observations strongly suggest that the melt compositions play an important role to control the partition coefficient between amphibole and glass. The REE become more enriched in

amphibole if the quenched glass is granodiorite or granite compared to the tonalitic glasses.

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