Appendix 1. Data sources

Calibrations

Muscovite. Agard et al. (2001); Ashworth and Evirgen (1985); Auzanneau et al. (2010); Bosse et al. (2002); Carswell et al. (1997, 2003); Castelli et al. (2007); Cetinkaplan et al. (2008); Clarke et al. (1997); Crowley (1990); Dale and Holland (2003); Davis and Whitney (2006); El-Shazly (1995); El-Shazly et al. (2001); Engvik and Andersen (2000); Faryad (1995); Fitzherbert et al. (2003, 2004); Foster et al. (2002); Fraser et al. (2000); Garcia-Casco et al. (2001); Gardien et al. (2000); Ghent et al. (2009); Groppo et al. (2007); Hermann and Spandler (2008); Hickmott and Spear (1992); House et al. (1997); Inger and Harris (1992); Keller et al. (2004, 2005); Kienast et al. (1991); Klemd et al. (2002); Kohn (2008); Kohn and Spear (1990); Kohn and Valley (1994); Kohn et al. (1993); Konzett and Hoinkes (1996); Krogh et al. (1994); Lang (1991); Lang and Gilotti (2007); Le Bayon et al. (2006); Léger and Ferry (1993); Lin et al. (2009); Liu et al. (2004, 2006); Massonne and Kopp (2005); Matsumoto et al. (2003); Menold et al. (2009); Meyre et al. (1999); Meza-Figueroa et al. (2003); Okay (2002); Palmeri et al. (2003); Patiño Douce (2005); Patrick (1995); Penniston-Dorland and Ferry (2006); Rolfo et al. (2004); Rolland et al. (2001); Rotzler et al. (1999); Schneider et al. (2004); Selverstone and Spear (1985); Selverstone et al. (1991); Sevigny and Ghent (1989); Shi and Wang (2006); Sisson et al. (1997); Smit et al. (2008); Song et al. (2003, 2007); Stípská et al. (2006); Verma et al. (2005); Wain et al. (2001); Walsh and Hacker (2004); Wang and Liou (1993); Warren and Waters (2006); Wei and Song (2008); Wei et al. (2009); White et al. (2001); Whitney and Ghent (1993); Zeh et al. (2005); Zhang and Liou (1997); Zhang et al. (2003, 2007, 2009); Zulbati (2008)

Biotite. Ashworth and Evirgen (1985); Auzanneau et al. (2010); Burgess et al. (1995); Conrad et al. (1988); Dusel-Bacon et al. (1995); Elvevold et al. (1994); Ferry (1981); Guidotti et al. (1977); Hansen (1992); Harris et al. (2004); Henry and Dokka (1992); Henry et al. (2005); Hoschek (2001); **{{AU: Reference not in list. Please add.}}** Jöns and Schenk (2004); Kohn (2008); Kohn and Spear (1990); Kohn and Valley (1994); Kohn et al. (1993); Konzett and Hoinkes (1996); Kühn et al. (2004); Labotka (1987); **{{AU: Reference not in list. Please add.}}** Lang (1991); Léger and Ferry (1993); Page et al. (2003); **{{AU: Reference not in list. Please add.}}** Palmeri et al. (2003); Patel et al. (1999); Patiño-Douce (2005); Patiño-Douce and Beard (1995); Selverstone and Spear (1985); Selverstone et al. (1984, 1991); Sevigny and Ghent (1989); Sommer et al. (2003); Spear (1982); Stípská et al. (2008); Thomson (1992); Tracy (1978); Tracy and Robinson (1988); Tucillo et al. (1992); **{{AU: Reference not in list. Please add.}}** Walsh and Hacker (2004); Whitney and Ghent (1993); Williams and Grambling (1990); Zeh et al. (2004).

Hornblende. Ague (2002); Allibone et al. (2009); Anovitz and Essene (1990); Coolen (1980); Daczko et al. (2001); Diaz Garcia et al. (1999); Ernst and Liu (1998); Faryad and Hoinkes (2003); Fazlnia et al. (2009); Janak et al. (2001); Klemd et al. (2002); Kohn and Spear (1990); Kohn et al. (1993); Konzett and Hoinkes (1996); Lee and Cho (2003); McGrew et al. (2000); Norlander et al. (2002); Oh et al. (2004); Sanchez-Vizcaino et al.

(2003); Selverstone et al. (1984); Sisson et al. (1997); Spear (1982); Storey et al. (2005); Surour (1995); Tsujimori et al. (2006); Valley et al. (2003); Zhang et al. (2010).

***a*(rt) calculations from GRIPS**

Muscovite. Abu El-Enen et al. (2003); Ahmed-Said and Tanner (2000); Anczkiewicz et al. (2007); Arenas et al. (1995); Argles (2000); Beppu and Okudaira (2006); Borghi et al. (2003); Brooijmans et al. (2003); Catlos et al. (2004); Dallmeyer et al. (1997); Dasgupta et al. (2004, 2009); Dusel-Bacon et al. (1995); Florence et al. (1993); Fraser et al. (2000); Gal and Ghent (1991); Goscombe and Hand (2000); Goswami et al. (2009); Hansen (1992); Harris et al. (2007); Henk et al. (1997); Hoisch and Simpson (1993); Hoisch et al. (2002); Hwang et al. (2001); Ilg et al. (1996); Johnston et al. (2007); Kim et al. (2005); Kohn et al. (1993); Likhanov et al. (2001, 2008, 2009); Mahato et al. (2008); Min and Cho (1998); Pattison and Vogl (2005); Swapp and Hollister (1991); Tracy (1978); Verma et al. (2005); Vry et al. (2004); Wang and Spear (1991); Whitney et al. (1996).

Biotite. Abu El-Enen (2008); Abu El-Enen et al. (2003, 2004, **{{AU: Reference not found in list. Please add.}}** 2011); Ahmed-Said and Tanner (2000); Anczkiewicz et al. (2007); Arenas et al. (1995); Argles (2000); Beppu and Okudaira (2006); Berman et al. (2010); Borghi et al. (2003); Brooijmans et al. (2003); Catlos et al. (2004); Dallmeyer (1997); Dasgupta et al. (2004, 2009); Dusel-Bacon et al. (1995); Florence et al. (1993); Fraser et al. (2000); Gal and Ghent (1991); Goscombe and Hand (2000); Goscombe et al. (1998); Goswami et al. (2009); Hansen (1992); Harris et al. (2007); Henk et al. (1997); Hoisch and Simpson (1993); Hoisch et al. (2002); Ilg et al. (1996); Johnston et al. (2007); Kawakami et al. (2007); Kim et al. (2005); Kohn and Spear (1993); Lee and Cho (1995); Likhanov et al. (2001, 2008, 2009); Mahato et al. (2008); Min and Cho (1998); Pattison and Vogl (2005); Pickett and Saleeby (1993); Swapp and Hollister (1991); Tracy (1978); Verma et al. (2005); Vry et al. (2004); Wang and Spear (1991); Whitney et al. (1996).

Hornblende. Balen et al. (2006); Ballevre et al. (2000); Barth and May (1992); Bisnath et al. (2008); Coolen (1980); De Paoli et al. (2009); Faryad (1999); Faryad and Hoinkes (2003); Galán and Marcos (2000); Garcia et al. (2003); Ghent and Stout (1986 2000); Glassley and Sorensen (1980); Gordon et al. (2010); Goscombe et al. (1998); Gupta et al. (2000); Klemd et al. (2002); Kohn et al. (1992, 1993); Lee and Cho (2003); Lexa et al. (2005); Liu et al. (2009); **{{AU: Reference not in list. Please add.}}** Mazur et al. (2005); Norlander et al. (2002); Oberc-Dziedzic et al. (2009); Percival (1983); Pitra et al. (2010); **{{AU: Reference not in list. Please add.}}** Kumar and Chacko (1994); Santosh et al. (2006); Selverstone et al. (1999); Stoddard (1985); Vogl (2003); Williams et al. (2000); Yang et al. (1998).

Appendix 2. Compositional projections for biotite and hornblende

Compositions may be readily projected from cations and vacancies to mole fractions using Excel's LINEST function. For hornblende, cations and vacancies are first formulated as: Si, Al+Fe^{3+}, Ti, Fe+Mg+Mn, Ca, Na+K, and alkali vacancy (= 16 – cation sum). For biotite, cations and vacancies are calculated as: Si, Al+Fe^{3+}, Ti, Fe+Mg+Mn, octahedral vacancy (= 7 – Si – Al – Fe^{3+} – Ti – Mg – Fe – Mn), Na, K, and alkali vacancy (= 1 – Na – K – Ca). Hornblende and biotite mole fractions are then calculated by regressing each composition vector (cv) against the following compositional definition matrices (cdm):

Amphibole	Si	Al	Ti	FM	Ca	Na+K	Vac
cmt	8	0	0	7	0	0	1
tr	8	0	0	5	2	0	1
edn	7	1	0	5	2	1	0
gl	8	2	0	3	0	2	1
TiAm	4	4	2	3	2	0	1
tsch	6	4	0	3	2	0	1

Biotite	Si	Al	Ti	FM	Oct vac	Na	K	Alk vac
phl	3	1	0	3	0	0	1	0
Tsbt	2	3	0	2	0	0	1	0
Tibt	3	1	1	1	1	0	1	0
cel	4	1	0	1	1	0	1	0
tlc	4	0	0	3	0	0	0	1
Nabt	3	1	0	3	0	1	0	0

That is, the 6 mole fractions are calculated by applying the array function "= LINEST(cv,cdm)" to the 6 target cells. **{{AU: should cel be celms?}}**

Examples of these projections are provided in the supplemental file.

Appendix 3. Compositional conversions among Ti-components in biotite

Commonly proposed Ti-components in biotite include Ti-oxy biotite [$K(Mg_2Ti)(AlSi_3)O_{10}(O)_2$], Ti-Tschermaks biotite [$K(Mg_2Ti)(Al_3Si)O_{10}(OH)_2$], and Ti-vacancy biotite [$K(\square MgTi)(AlSi_3)O_{10}(OH)_2$]. These are all interconvertible with accepted exchanges as follows (relating Ti-oxy and Ti-vacancy biotite to an intermediary Ti-Tschermaks biotite)

$K(Mg_2Ti)(AlSi_3)O_{10}(O)_2 + 2(AlAl)(MgSi)_{-1} = KAl_2TiAl_3SiO_{10}(O)_2$ (Tschermak's exchange)

$KAl_2TiAl_3SiO_{10}(O)_2+2Fe^{3+}(Al)_{-1} = K(Fe^{3+}{}_2Ti)(Al_3Si)O_{10}(O)_2$ (Fe^{3+}-Al exchange)

$K(Fe^{3+}{}_2Ti)(Al_3Si)O_{10}(O)_2+2Fe^{2+}H(Fe^{3+})_{-1} = K(Fe^{2+}{}_2Ti)(Al_3Si)O_{10}(OH)_2$ (Fe-oxy exchange)

$K(Fe^{2+}{}_2Ti)(Al_3Si)O_{10}(OH)_2+2MgFe_{-1} = K(Mg_2Ti)(Al_3Si)O_{10}(OH)_2$ (Fe-Mg exchange)

$K_2(\square_2Mg_2Ti_2)(Al_2Si_6)O_{20}(OH)_4+2(AlAl)(MgSi)_{-1} = K_2(\square_2Al_2Ti_2)(Al_4Si_4)O_{20}(OH)_4$ (Tschermak's exchange)

$K_2(\square_2Al_2Ti_2)(Al_4Si_4)O_{20}(OH)_4+Mg_3(\square Al_2)_{-1} = K_2(\square Mg_3Ti_2)(Al_4Si_4)O_{20}(OH)_4$ (dioct.-trioctahedral exchange)

$K_2(\square Mg_3Ti_2)(Al_4Si_4)O_{20}(OH)_4+2(AlAl)(MgSi)_{-1} = K_2(\square Al_2MgTi_2)(Al_4Si_4)O_{20}(OH)_4$ (Tschermak's exchange)

$K_2(\square Al_2MgTi_2)(Al_4Si_4)O_{20}(OH)_4+Mg_3(\square Al_2)_{-1} = K_2(Mg_4Ti_2)(Al_6Si_2)O_{20}(OH)_4$ (dioct.-trioctahedral exchange)

The applied exchanges are known to occur in micas (and amphibole), justifying our choice of end-member components. Alternative end-member components could be chosen that include Ti-Tschermak's biotite or Ti-vacancy biotite that would lead to identical activity expressions and regression results.

A similar rationale applies to Ti-components of muscovite and hornblende

Appendix 4. *a*(rt) calculations

Pressure-temperature conditions of equilibration were estimated using the garnet-hornblende (Graham and Powell 1984), plagioclase-hornblende (Holland and Blundy 1994) or garnet-biotite [Ferry and Spear (1978), with Berman (1990), garnet activity model] thermometers and the garnet-plagioclase-hornblende-quartz (GPHQ; Kohn and Spear 1990) and garnet-plagioclase-muscovite-biotite (GPMB; Hoisch 1990) barometers. We cross-checked these *P-T* estimates against the Thermocalc database (Holland and Powell 1990, 1998) using the "optimal" *P-T* method of Powell and Holland (1994) and found no systematic offset between results. We also found that differences in calculated temperature and pressure with the two methods were correlated with a slope nearly identical to the GRIPS equilibrium (within 1 bar/°C). Thus, different thermobarometric methods would not produce significant differences in *a*(rt). A ~1 kbar difference occurs between the original GRIPS calibration (Bohlen and Liotta 1986) and the Thermocalc database as well as between calculated pressures in natural rocks using GRIPS and the garnet-plagioclase-aluminosilicate-quartz equilibrium (GASP; Wu and Zhao 2006). That is, the same ~1 kbar pressure difference is observed between estimated end-member reaction positions (Bohlen and Liotta vs. Thermocalc), and calculated pressures in natural rocks (GRIPS vs. GASP). Hoisch (1990) based his calibrations on the GASP reaction (Koziol and Newton 1988). **{{AU: Reference not in list. Please add.}}** So for consistency we use the Thermocalc calibration of GRIPS for estimating *a*(rt), rather than

Bohlen and Liotta (1986). The Bohlen and Liotta calibration yields estimated a(rt) values c. 20% lower, e.g., 0.1 lower at a(rt) = 0.5, 0.2 lower at a(rt) = 1.0, etc.