Revision 1

Supplementary Information

Re-examined heterotype solid solution between calcite and strontianite and Ca-Sr fluid-carbonate distribution: an experimental study in the system $CaCO_3$–$SrCO_3$–H_2O at 0.5–5 kbar and 600°C

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Table S1: Starting materials, initial weights, and experimental conditions for preparing metastable Sr-Arg educt material for type III.

	U28	**U29**	**U30**
H_2O (L)	0.500	0.500	0.500
Temperature (°C)	80–90	80–90	80–90
$SrCl_2$*6$H_2$0 (g L^{-1})	17.06	23.46	12.8
$CaCl_2$*2H_2O (g L^{-1})	14.12	10.58	16.46
Urea (g L^{-1})	12.00	12.00	12.00
$SrCl_2$*6$H_2$0 (mol L^{-1})	0.032	0.044	0.024
$CaCl_2$*2H_2O (mol L^{-1})	0.048	0.036	0.056
Urea (mol L^{-1})	0.10	0.10	0.10
x_{Sr} (initial)	0.40	0.55	0.30
XRD			
Major phase	Sr-aragonite	Sr-aragonite	Sr-aragonite
Weight fraction (wt-%)	100	100	100
Other phases	<LOD	< LOD	< LOD
SEM			
Major phase	Sr-aragonite	Sr-aragonite	Sr-aragonite
$x_{Sr}(Arg_{ss})$ [a]	0.24	0.37	0.24

[a] arithmetic mean of three EDX measurements

Table S2: Refined lattice parameters. Estimated six-fold standard errors for the Rietveld refinement are given in parentheses.

	orthorhombic	trigonal	orthorhombic			trigonal	
	(mol% $SrCO_3$)		*a* (Å)	*b* (Å)	*c* (Å)	*a* (Å)	*c* (Å)
P13	56.7±4.5	25.9±2.6	5.0458 (6)	8.2110 (14)	5.9176 (8)	5.0096 (8)	17.259 (3)
P17	49.4±1.3	24.6±2.6	5.0367 (5)	8.1863 (10)	5.8986 (8)	4.9986 (5)	17.176 (2)
P18	44.6±3.8	22.3±2.2	5.0316 (6)	8.1710 (10)	5.8871 (8)	4.9996 (5)	17.211 (2)
P19	41.8±2.7	21.4±2.5	5.0259 (6)	8.1495 (10)	5.8747 (7)	5.0016 (12)	17.177 (4)
P20	38.8±2.8	18.4±0.5	5.0216 (8)	8.1352 (10)	5.8662 (10)	4.9980 (13)	17.161 (4)
P21	54.2±2.0	26.0±1.0	5.0446 (9)	8.2055 (11)	5.9137 (16)	5.0063 (36)	17.213 (15)
P22	53.1±1.7	24.7±1.2	5.0414 (4)	8.1967 (10)	5.9085 (13)	5.0023 (21)	17.205 (8)

Table S3: Thermodynamic data for modelling P-x relation at 600°C based on a symmetric regular solution model.

h	=standard enthalpy	(kJ mol^{-1})
s	=standard entropy	(kJ mol^{-1} K^{-1})
v	=molar volume	(J MPa^{-1} mol^{-1})
α	=thermal expansion	(K^{-1})
β	=compressibility	(MPa^{-1})
Cp	=heat capacity as function of temperature	(J K^{-1} mol^{-1})

Aragonite

h	$= -1205554$	(Gottschalk 1997)
s	$= 91.341$	(Gottschalk 1997)
v	$= 34.150$	(Robie et al. 1978)
α	$= 72.7\times10^{-6}$	(Gottschalk 1997)
β	$= 15.5\times10^{-6}$	(Gottschalk 1997)
C_{P1}[a]	$= 81.533+4.5673\times10^{-2}\times T-1.1405\times10^{6}\times T^{-2}$	(Robie et al. 1978)
C_{P2}[b]	$= 166.61-14.994\times10^{2}\times T^{-0.5}+5.499\times10^{7}\times T^{-3}$	(Bermann 1985)

[a] for T< 600 K; [b] for T>600 K

Strontianite

h	= -1231400	(Kiselava 1994)
s	= 97.2	(Nordstrom 2013)
v	= 38.981	(Kiselava 1994)
α	$= 58.3\times10^{-6}$	(Gottschalk 1997)
β	$= 16.13\times10^{-6}$	(Gottschalk 1997)
C_P	$= -81.596+0.10754\times T+3.1677\times10^{3}\times T^{-0.5}-1.3914\times10^{9}\times T^{-3}$	(Robie et al.)

Calcite

h	$= -1206686$	(Gottschalk 1997)
s	$= 91.487$	(Gottschalk 1997)

v	= 36.89	(Holland&Powell 2011)
α	$= 28.5\times10^{-6}$	(Gottschalk 1997)
β	$= 13.7\times10^{-6}$	(Gottschalk 1997)
C_P	$= 99.715+2.6920\times10^{-2}\times T-2.1576\times10^{6}\times T^{-2}$	(Robie et al. 1978)
C_p	$= 193.24-2.0409\times10^{3}\times T^{-0.5}+19.946\times10^{7}\times T^{-3}$	(Bermann 1985)

Trigonal $SrCO_3$

v	= 39.7*	(this study)
α	$= 28.5\times10^{-6}$	(Gottschalk 1997)
β	$= 13.7\times10^{-6}$	(Gottschalk 1997)

*Molar volume of the trigonal Sr endmember was calculated from XRD measurements of this study and yielded 39.7 $cm^3\ mol^{-1}$.

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