

Electronic Appendix to:

C.A.J. Appelo, Principles, caveats and improvements in databases for calculating hydrogeochemical reactions in saline waters from 0 - 200 °C and 1 - 1000 atm.

This appendix has figures showing experimental solubilities and PHREEQC calculations, using the Pitzer interaction coefficients given in Table 1, and the temperature dependent log K 's in Table 2 of the paper. The PHREEQC input files can be downloaded as zip from:

http://www.hydrochemistry.eu/pub/pitzer_db/appendix.zip

Extract the file in your computer. The main directory contains pitzer.dat and input files that calculate the figures presented by Pabalan and Pitzer, 1987. Input files for other minerals and CO₂ gas are in sub-directories.

It is easiest to run the files with Notepad++ adapted for PHREEQC. Download from:

<http://www.hydrochemistry.eu/ph3/phreeqc3.Installer.exe>

and install in your computer. In Notepad++, open a file (Ctrl+O), and press Ctrl+F6 to start the PHREEQC calculations.

Table 3. List of figures with mineral solubilities as a function of T , P and solution composition.

Mineral solubility in water or aqueous solution	Temp °C	Pressure / atm	Figure
halite (NaCl)	0 - 300	1 - P_{sat}	A1
sylvite (KCl)	10 - 300	1 - P_{sat}	A2
bischofite ($MgCl_2 \cdot 6H_2O$), $MgCl_2 \cdot 2H_2O$, $MgCl_2 \cdot 4H_2O$	0 - 200	1 - P_{sat}	A3
mirabilite ($Na_2SO_4 \cdot 10H_2O$), thenardite (Na_2SO_4)	0 - 220	1 - P_{sat}	A4
arcanite (K_2SO_4)	0 - 210	1 - P_{sat}	A5
epsomite ($MgSO_4 \cdot 7H_2O$), hexahydrite ($MgSO_4 \cdot 6H_2O$), kieserite ($MgSO_4 \cdot H_2O$)	0 - 200	1 - P_{sat}	A6
halite (NaCl), sylvite (KCl) in Na/K-Cl solutions	0 - 200	1 - P_{sat}	A7
carnallite ($KMgCl_3 \cdot H_2O$) in K/Mg-Cl solutions	0 - 75	1	A8
gypsum ($CaSO_4 \cdot 2H_2O$)	0 - 95	1	A9
gypsum ($CaSO_4 \cdot 2H_2O$) in NaCl solutions	0.5 - 95	1	A10
gypsum ($CaSO_4 \cdot 2H_2O$), mirabilite ($Na_2SO_4 \cdot 10H_2O$), glauberite ($Na_2Ca(SO_4)_2$) and thenardite (Na_2SO_4) in Na_2SO_4 solutions	25 - 100	1	A11
gypsum ($CaSO_4 \cdot 2H_2O$) and anhydrite ($CaSO_4$)	30 - 160	1 - 1000	A12
anhydrite ($CaSO_4$) in NaCl solutions	100 - 200	1 - P_{sat}	A13
anhydrite ($CaSO_4$) in NaCl solutions	100 - 200	1 - 987	A14
anhydrite ($CaSO_4$) and glauberite ($Na_2Ca(SO_4)_2$) in Na_2SO_4 solutions	100 - 200	1 - P_{sat}	A15
anhydrite ($CaSO_4$), Goergeyite ($K_2Ca_5(SO_4)_6H_2O$) and syngenite ($K_2Ca(SO_4)_2 \cdot H_2O$) in K_2SO_4 solutions	100 - 200	1 - P_{sat}	A16
amorphous silica ($SiO_2(a)$) in NaCl solutions	25 - 300	1 - P_{sat}	A17
amorphous silica ($SiO_2(a)$) in Na_2SO_4 solutions	25 - 300	1 - P_{sat}	A18
amorphous silica ($SiO_2(a)$) in $MgCl_2$ solutions	25 - 300	1 - P_{sat}	A19
amorphous silica ($SiO_2(a)$) in $MgSO_4$ solutions	25 - 250	1 - P_{sat}	A20
amorphous silica ($SiO_2(a)$) in Li-Cl/ NO_3 solutions	25	1	A21
amorphous silica ($SiO_2(a)$) in K-Cl/ NO_3 solutions	25	1	A22
amorphous silica ($SiO_2(a)$) in $CaCl_2$ solutions	25	1	A23
barite ($BaSO_4$) in NaCl solutions	1 - 250	1 - P_{sat}	A24
barite ($BaSO_4$) in NaCl solutions	150 - 250	493	A25
calcite ($CaCO_3$) in NaCl solutions	10 - 60	1	A26
calcite ($CaCO_3$) in 3 M NaCl, variable CO_2	200	580	A27
calcite ($CaCO_3$) in NaCl solutions	120 - 260	12	A28
calcite ($CaCO_3$) at 1 bar CO_2 pressure	0 - 300	1 - ($P_{sat}+1$)	A29
calcite ($CaCO_3$) in NaCl solutions at 1 bar CO_2 pressure	0 - 250	1 - 1450	A30
CO_2 gas	25 - 100	1 - 710	A31
CO_2 gas in 1 and 6 M NaCl solution	25 - 300	35 - 200	A32
CO_2 gas in 4 M NaCl solution	80 - 180	9 - 95	A33
CO_2 gas in Na_2SO_4 solutions	140	12 - 96	A34
CO_2 gas in 2.3 M $CaCl_2$ solution	75 - 120	22 - 655	A35
CO_2 fugacity coefficients	0 - 300	99 - 987	A36

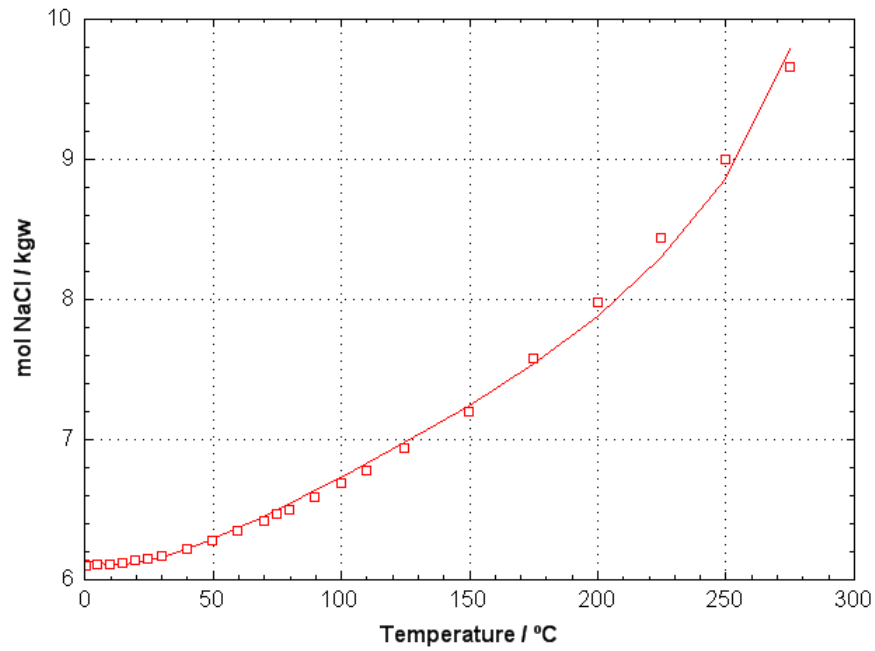


Figure A1. Halite (NaCl) solubility as a function of temperature. Data points from Pabalan and Pitzer, 1987; Clarke and Glew, 1985. File Halite.phr

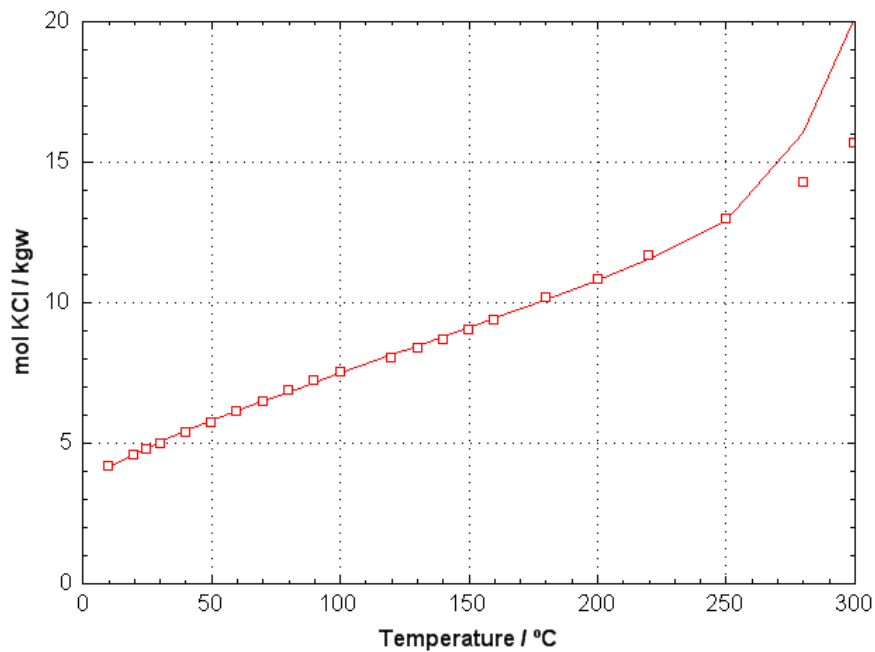


Figure A2. Sylvite (KCl) solubility as a function of temperature. Data points from Pabalan and Pitzer, 1987. File Sylvite.phr

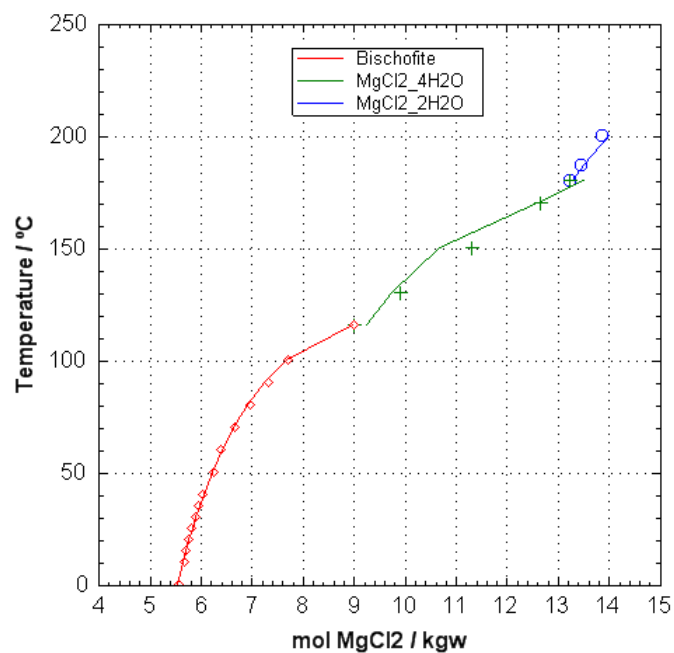


Figure A3. Solubility of MgCl_2 -hydrates. Data points from Pabalan and Pitzer, 1987. File $\text{MgCl}_2.\text{phr}$

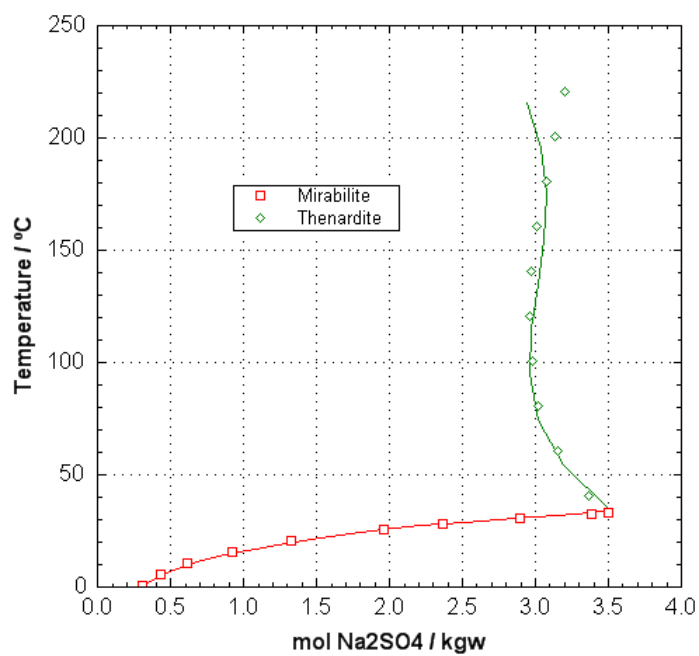


Figure A4. Solubility of Na_2SO_4 -(an)hydrate. Data points from Pabalan and Pitzer, 1987. File $\text{Na}_2\text{SO}_4.\text{phr}$

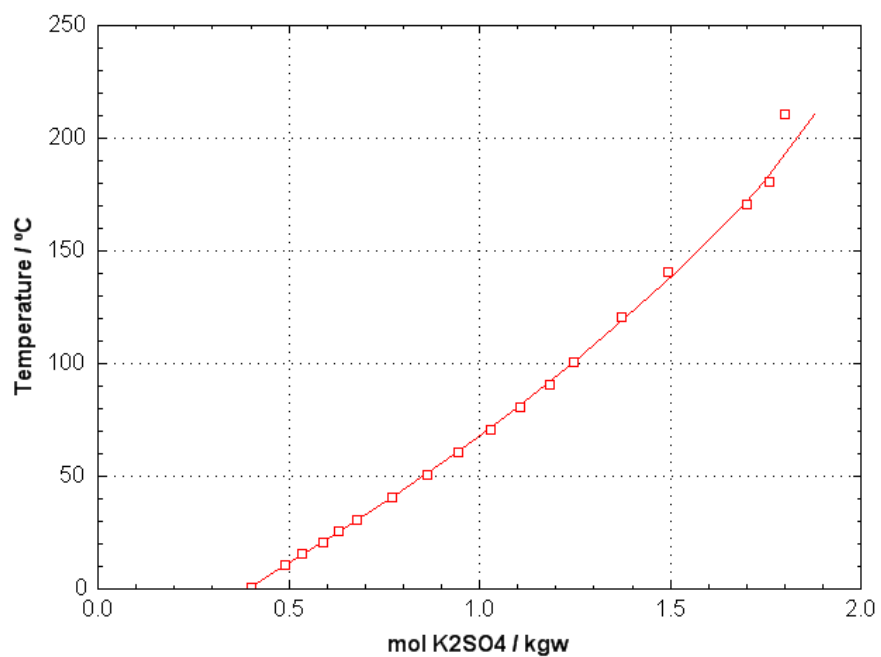


Figure A5. Solubility of arcanite (K_2SO_4). Data points from Pabalan and Pitzer, 1987. File K2SO4.phr

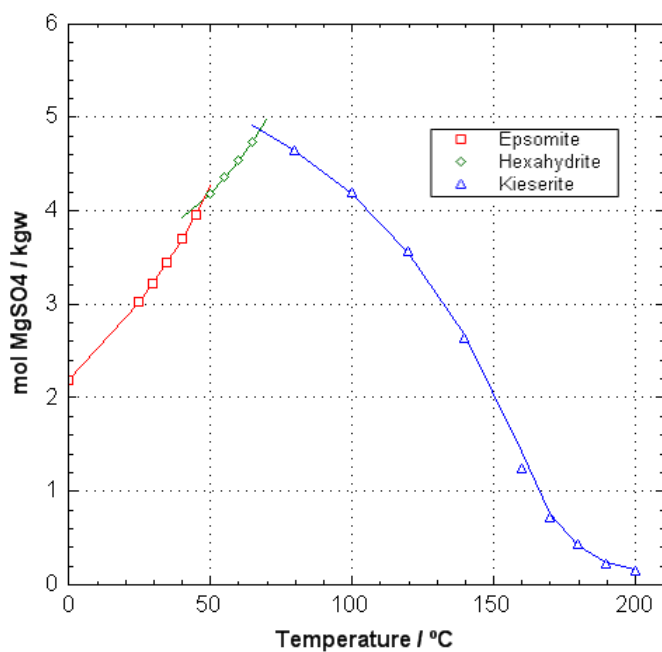


Figure A6. Solubility of $MgSO_4$ -hydrates. Data points from Pabalan and Pitzer, 1987. File MgSO4.phr

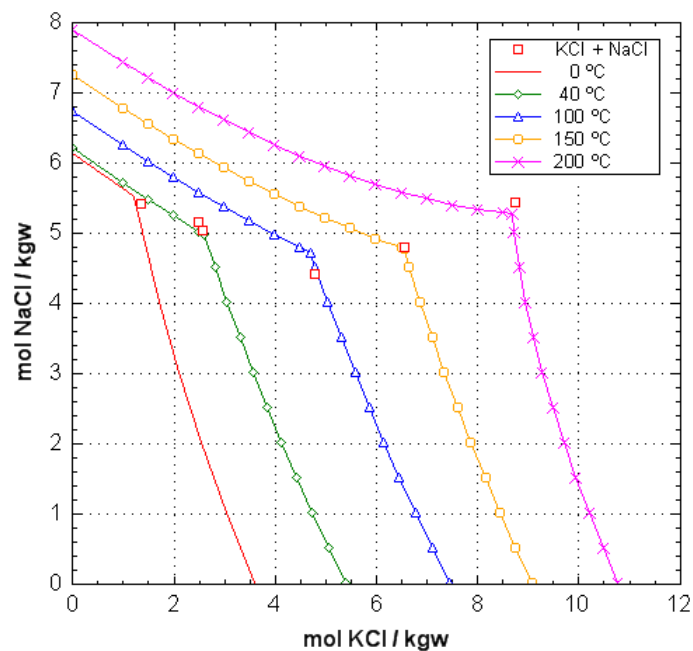


Figure A7. Mutual influence of NaCl and KCl solutions on halite and sylvite solubilities, with triple points KCl + NaCl + H₂O from Pabalan and Pitzer, 1987. File NaKCl.phr

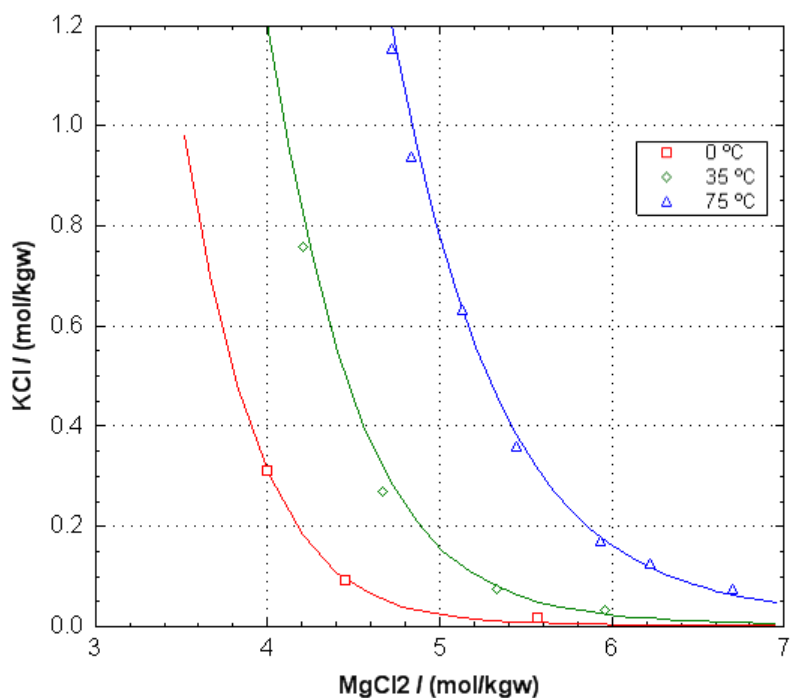


Figure A8. Solubility of carnallite (KMgCl₃·H₂O). Data points from Pabalan and Pitzer, 1987. File Carnallite.phr

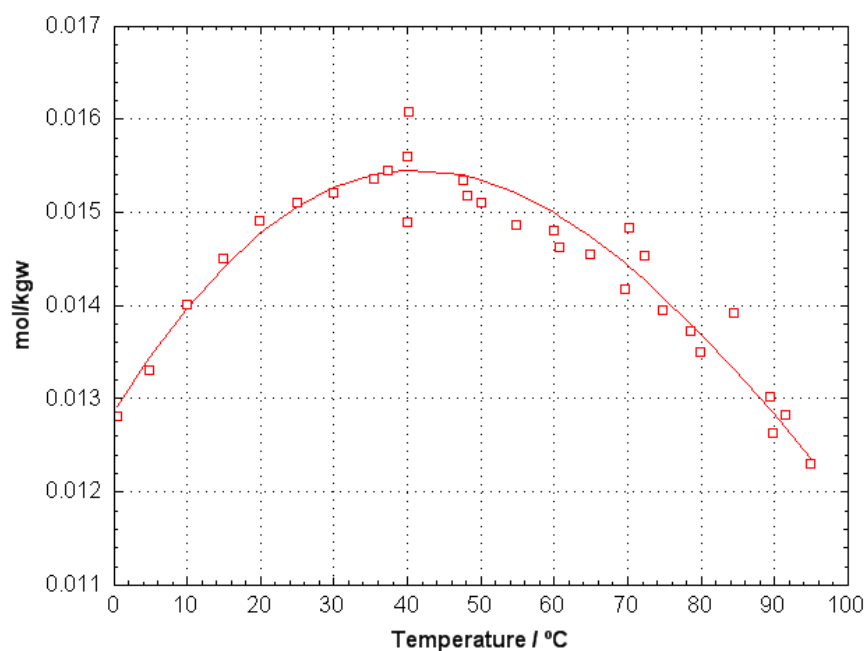


Figure A9. Solubility of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). Data points from Marshall and Slusher, 1966; Blount and Dickson, 1969. File gypsum.phr

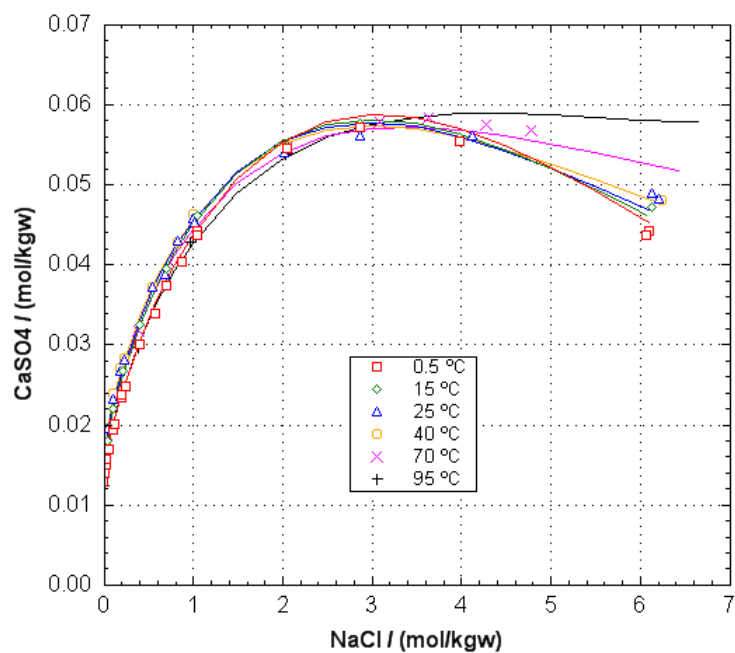


Figure A10. Solubility of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) in NaCl solutions. Data points from Marshall and Slusher, 1966. File gyps_NaCl.phr

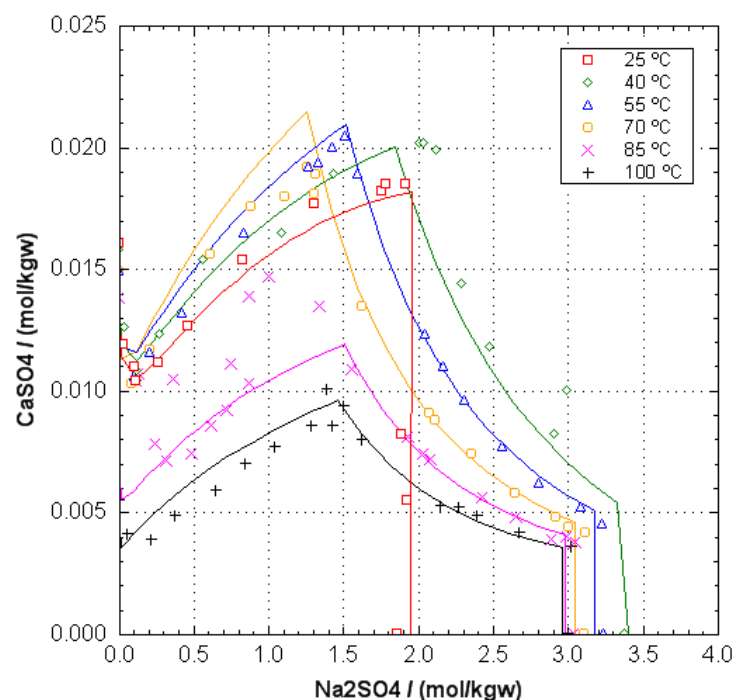


Figure A11. Solubility of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) in Na_2SO_4 solutions. Data points from Block and Waters, 1968. File gyps_Na2SO4.phr. The concentration decrease of CaSO_4 at Na_2SO_4 concentrations above 1 M is due to mirabilite precipitation (25°C), or glauberite and thenardite precipitation at higher temperatures. At 85 and 100°C, and possibly at 70°C, gypsum transforms into anhydrite.

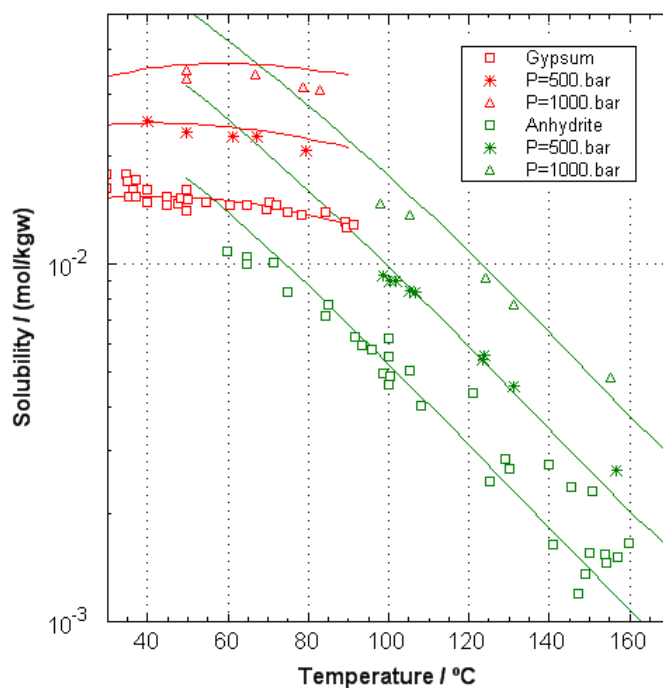


Figure A12. Solubility of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and anhydrite (CaSO_4) as a function of temperature and pressure. Data points from Blount and Dickson, 1973. File gypsum_P.phr.

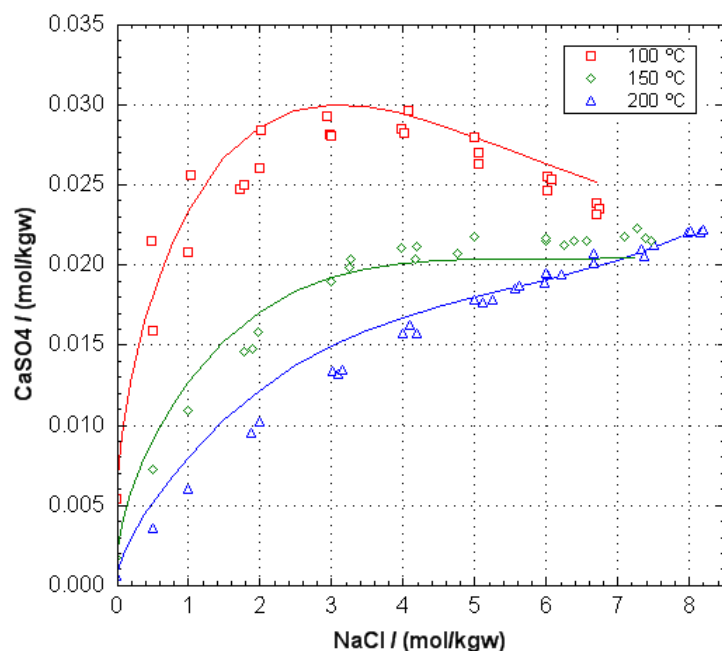


Figure A13. Solubility of anhydrite (CaSO_4) in NaCl solutions. Data points from Block and Waters, 1968; Blount and Dickson, 1969; Freyer and Voigt, 2004. File anhy_NaCl.phr.

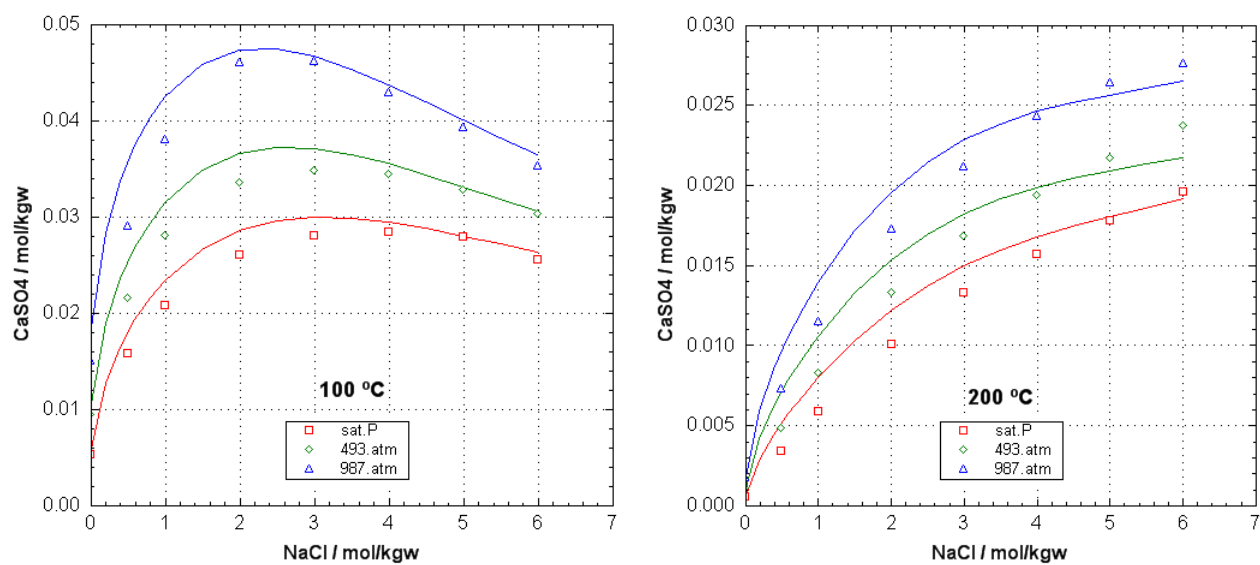


Figure A14. Solubility of anhydrite (CaSO_4) in NaCl solutions as a function of pressure at 100 °C and 200 °C. Data points from the summary table in Blount and Dickson, 1969. File anhy_P_NaCl.phr.

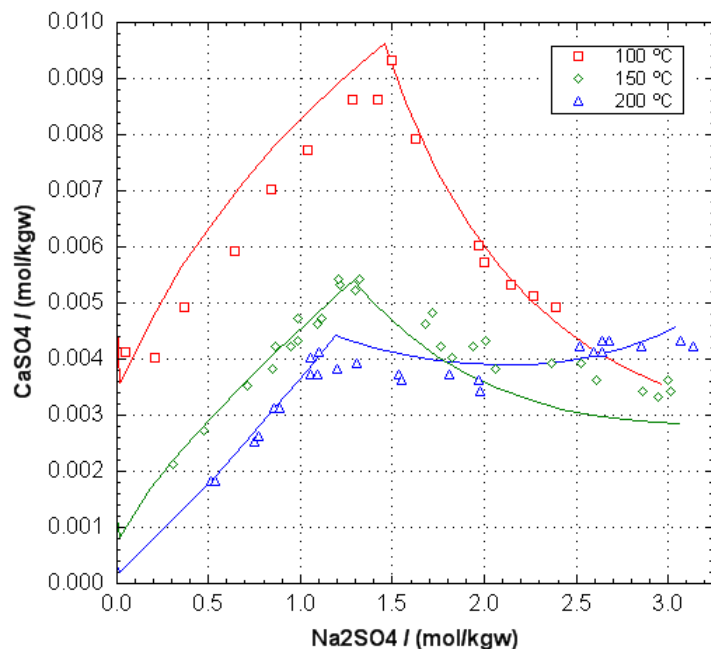


Figure A15. Solubility of anhydrite (CaSO_4) in Na_2SO_4 solutions. Data points from Freyer and Voigt, 2004. The decrease of the CaSO_4 concentration at Na_2SO_4 concentrations above 1 M is due to glauberite precipitation. File anhy_ Na_2SO_4 .phr.

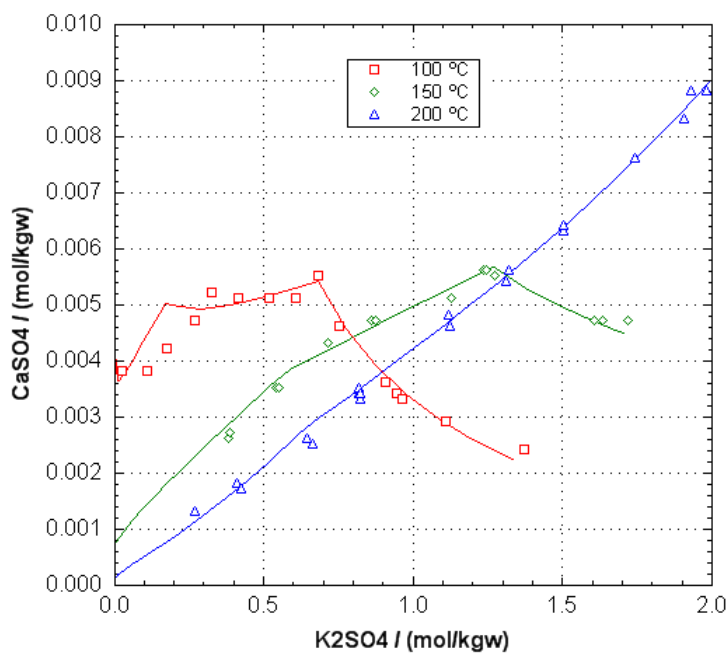


Figure A16. Solubility of anhydrite (CaSO_4) in K_2SO_4 solutions. Data points from Freyer and Voigt, 2004. The breaks in the concentration lines result from precipitation of Goergeyite ($\text{K}_2\text{Ca}_5(\text{SO}_4)_6\text{H}_2\text{O}$) and, at 100 and 150 °C, Syngenite ($\text{K}_2\text{Ca}(\text{SO}_4)_2\cdot\text{H}_2\text{O}$). File anhy_ K_2SO_4 .phr.

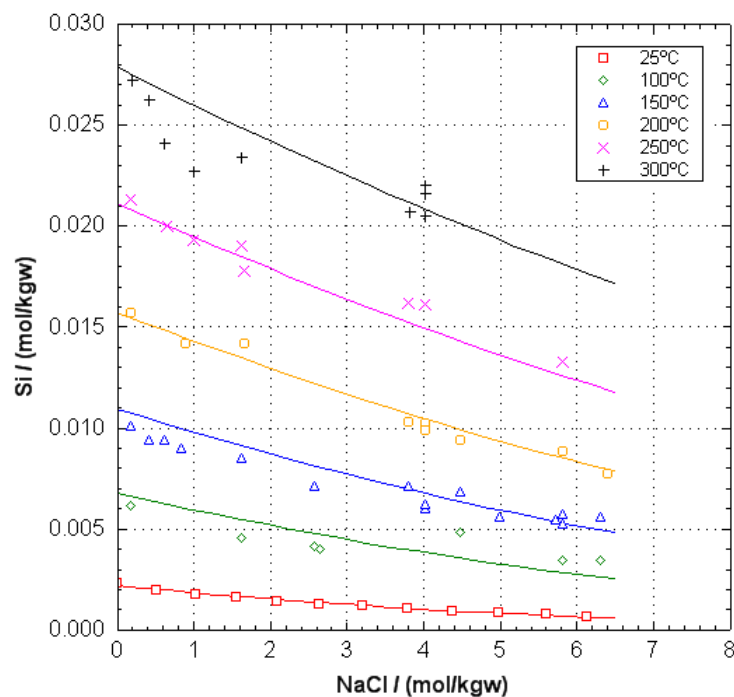


Figure A17. Solubility of amorphous silica ($\text{SiO}_2(\text{a})$) in NaCl solutions. Data points from Marshall and Warakomski, 1980, and Chen and Marshall, 1982. File SiO2_NaCl.phr.

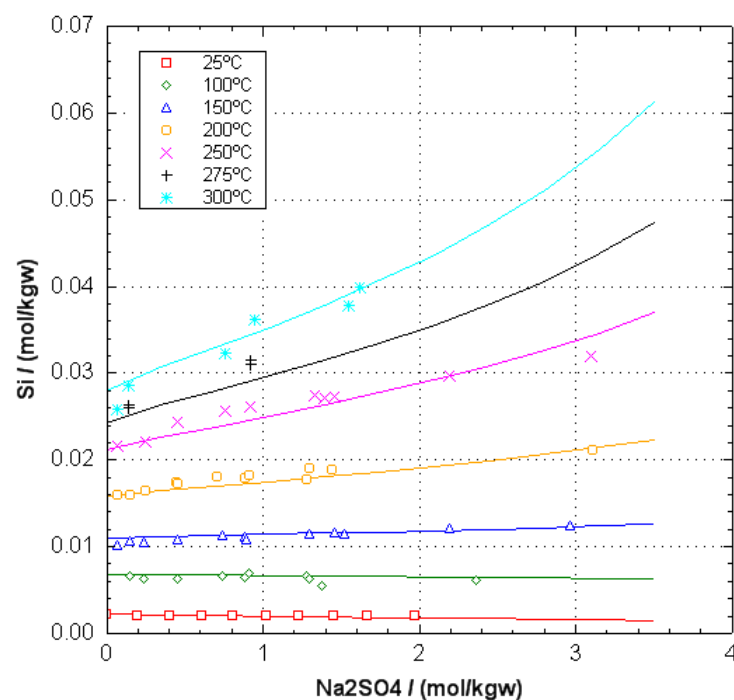


Figure A18. Solubility of amorphous silica ($\text{SiO}_2(\text{a})$) in Na_2SO_4 solutions. Data points from Marshall and Warakomski, 1980, and Chen and Marshall, 1982. File SiO2_Na2SO4.phr.

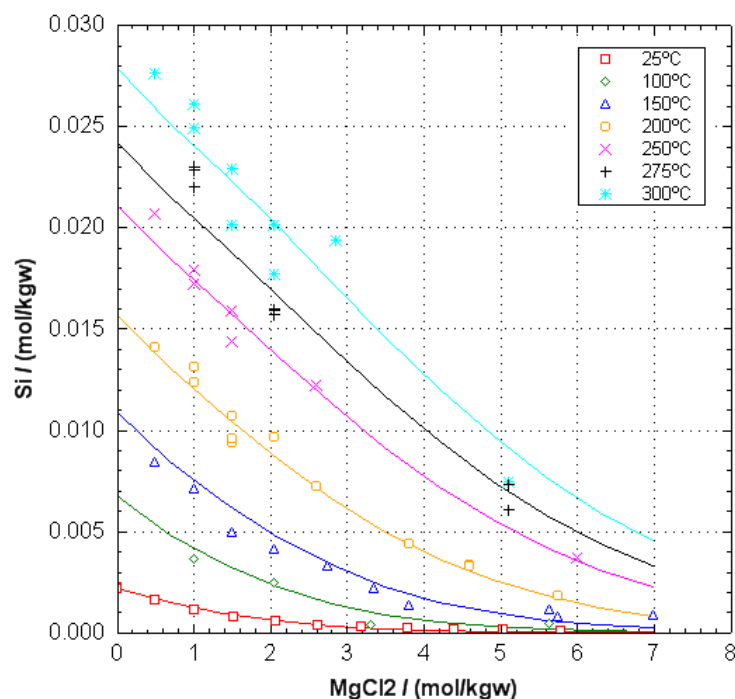


Figure A19. Solubility of amorphous silica ($\text{SiO}_2(\text{a})$) in MgCl_2 solutions. Data points from Marshall and Warakomski, 1980, and Chen and Marshall, 1982. File $\text{SiO}_2\text{-MgCl}_2.\text{phr}$.

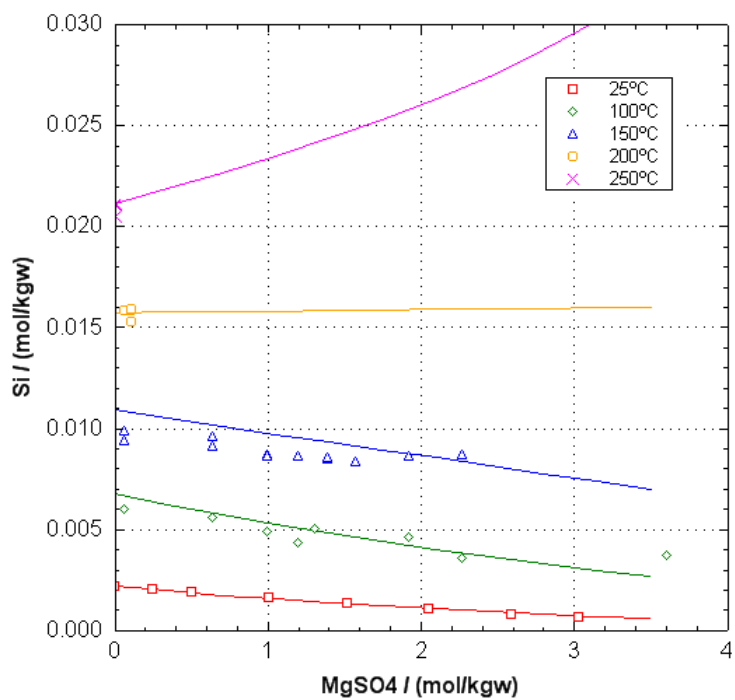


Figure A20. Solubility of amorphous silica ($\text{SiO}_2(\text{a})$) in MgSO_4 solutions. Data points from Marshall and Warakomski, 1980, and Chen and Marshall, 1982. File $\text{SiO}_2\text{-MgSO}_4.\text{phr}$.

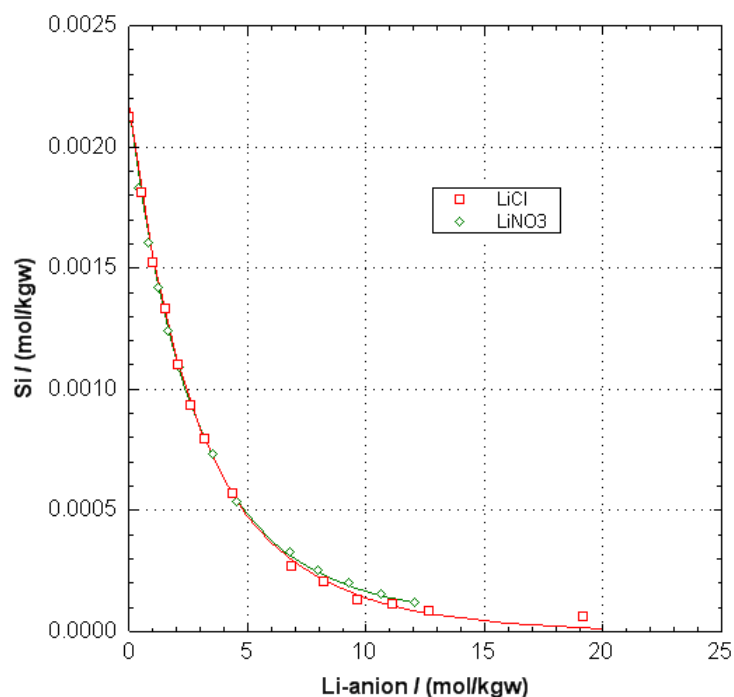


Figure A21. Solubility of amorphous silica ($\text{SiO}_2(\text{a})$) in LiCl and LiNO_3 solutions at 25°C. Data points from Marshall and Warakomski, 1980. File SiO2_Li.phr.

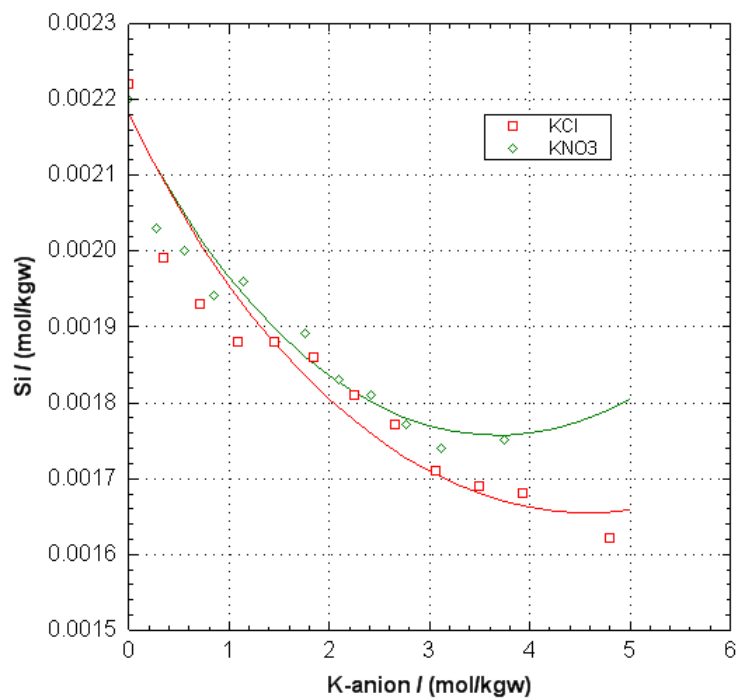


Figure A22. Solubility of amorphous silica ($\text{SiO}_2(\text{a})$) in KCl and KNO_3 solutions at 25°C. Data points from Marshall and Warakomski, 1980. File SiO2_K.phr.

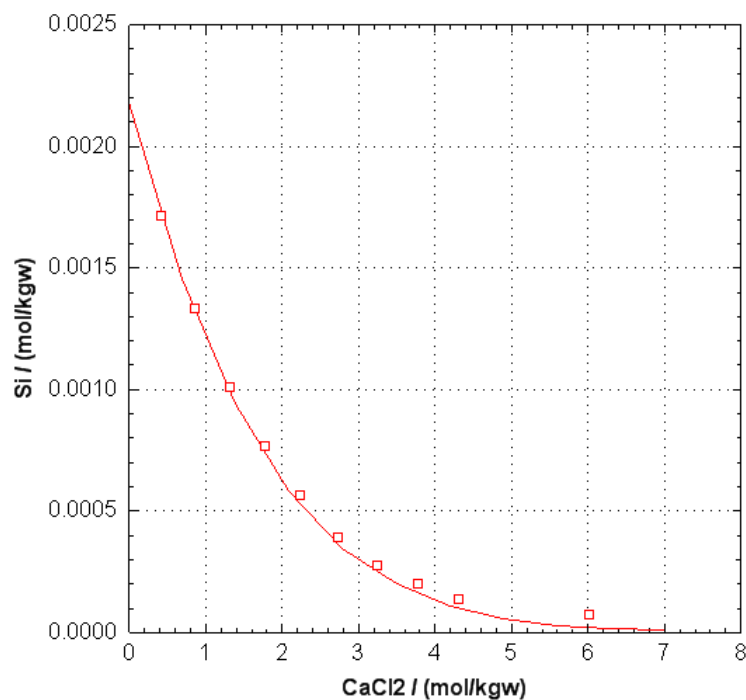


Figure A23. Solubility of amorphous silica ($\text{SiO}_2(\text{a})$) in CaCl_2 solutions at 25°C . Data points from Marshall and Warakomski, 1980. File `SiO2_CaCl2.phr`.

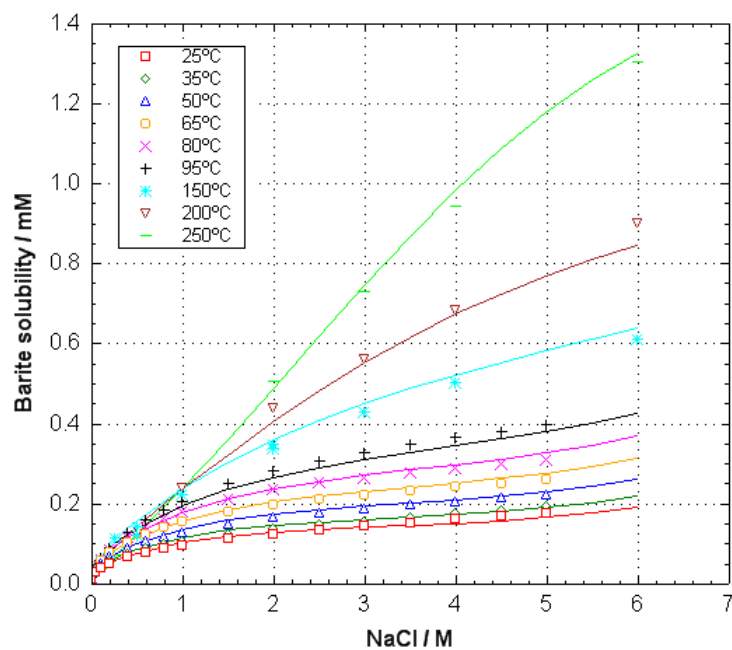


Figure A24. Solubility of barite (BaSO_4) in NaCl solutions. Data points $< 95^\circ\text{C}$ from Templeton, 1960, $> 100^\circ\text{C}$ from Uchameyshvili et al., 1966, and the summary table in Blount, 1977. File `Barite_NaCl.phr`.

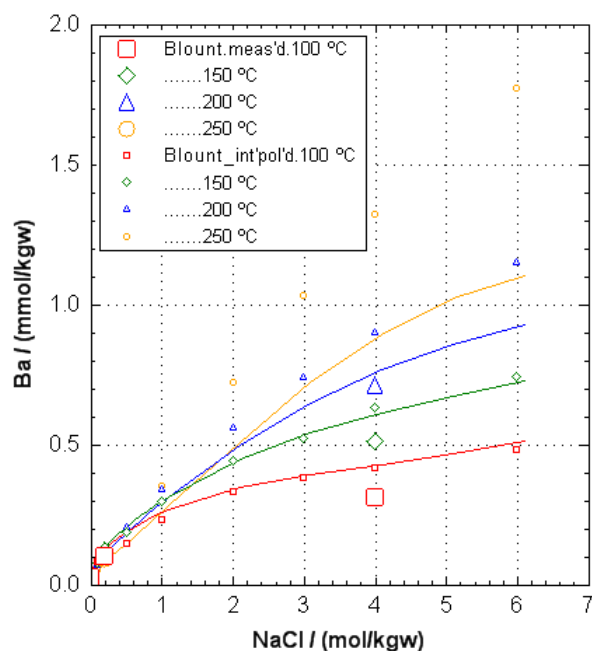


Figure A25. Solubility of barite (BaSO_4) in NaCl solutions at 500 bar. Measured (large symbols) and interpolated (small symbols) data from Blount, 1977. File Barite_500.phr.

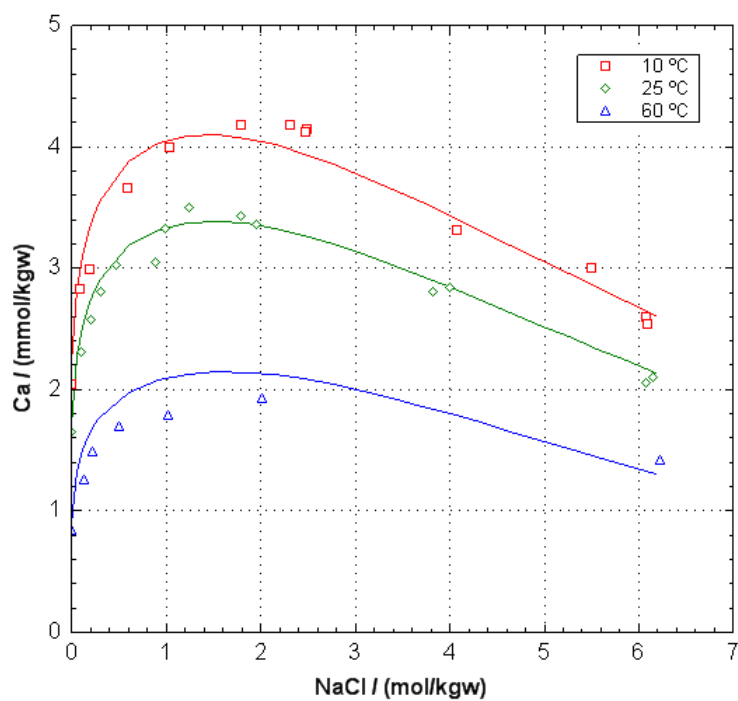


Figure A26. Solubility of calcite (CaCO_3) in NaCl solutions at about 0.01 atm CO_2 pressure. Measured data from Wolf et al., 1989. File cc_Wolf.phr.

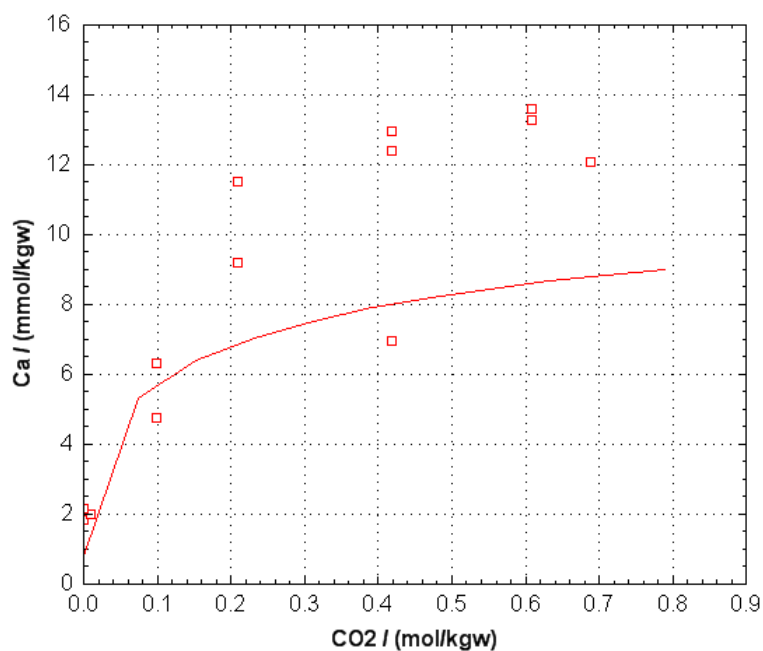


Figure A27. Solubility of calcite (CaCO_3) at 200 °C, 580 atm pressure in 3 M NaCl as a function of the CO_2 concentration. Measured data from Malinin and Kanukov, 1971. File cc_Malin.phr.

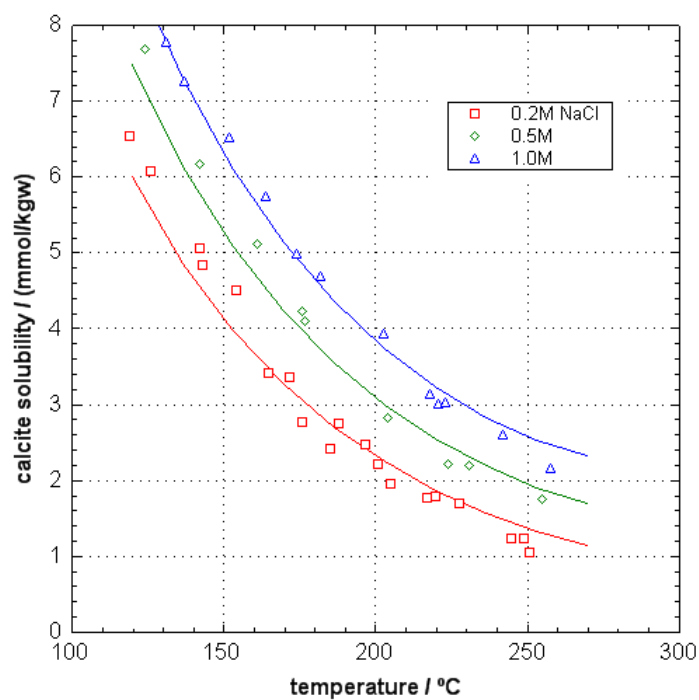


Figure A28. Solubility of calcite (CaCO_3) in NaCl solutions at 12 bar CO_2 pressure. Measured data from Ellis, 1963. File cc_Ellis.phr.

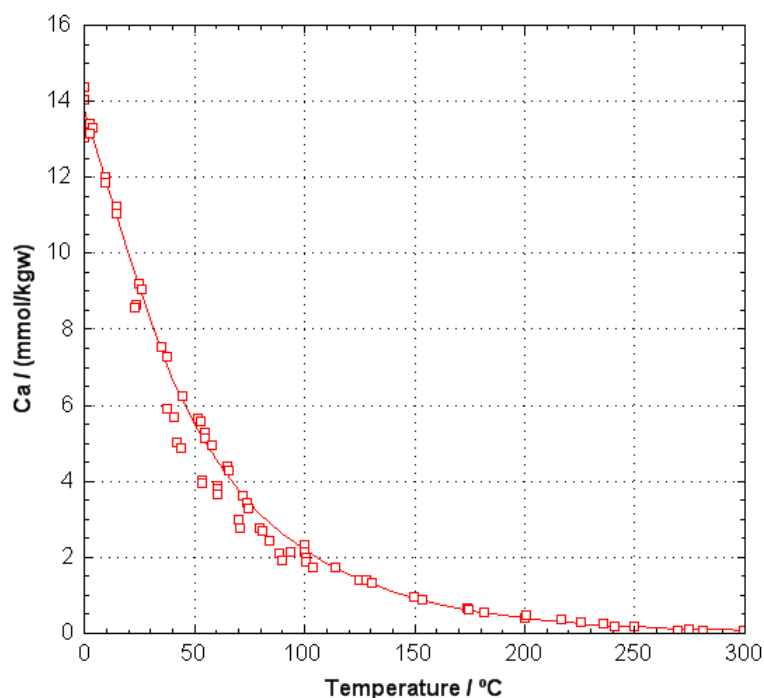


Figure A29. Solubility of calcite (CaCO_3) as a function of temperature at 1 bar CO_2 pressure. Measured data from Miller, 1952; Ellis, 1959; Plummer and Busenberg, 1982. File cc_1barCO2.phr.

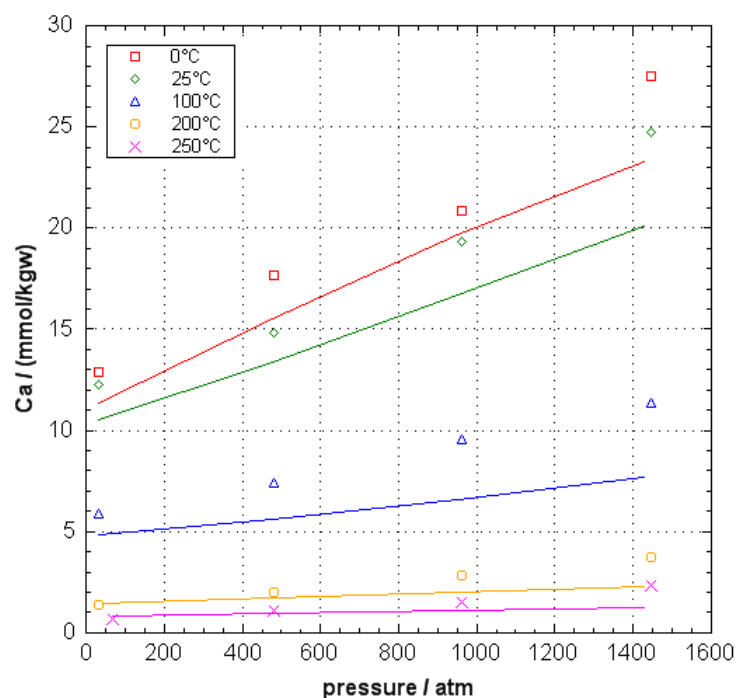


Figure A30. Solubility of calcite (CaCO_3) in 0.1 M NaCl as a function of temperature and pressure at 1 bar CO_2 pressure. Measured data from Shi et al., 2013. File cc_Shi.phr. Solubilities at 4 M NaCl and in a brine are also calculated when the file is run.

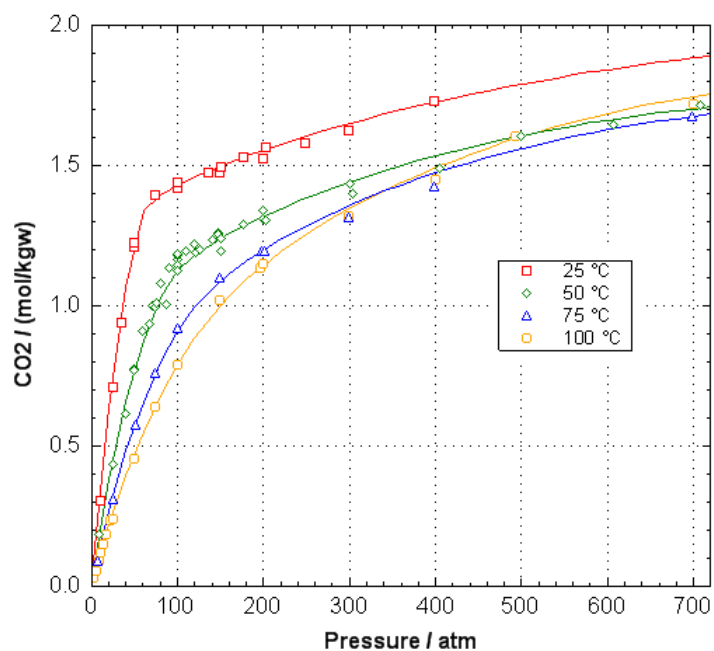


Figure A31. Solubility of CO₂ gas in water. Measured data from Wiebe and Gaddy, 1939, 1940; King et al., 1992; Takenouchi and Kennedy, 1964. File CO2_conc_PR_IS.phr.

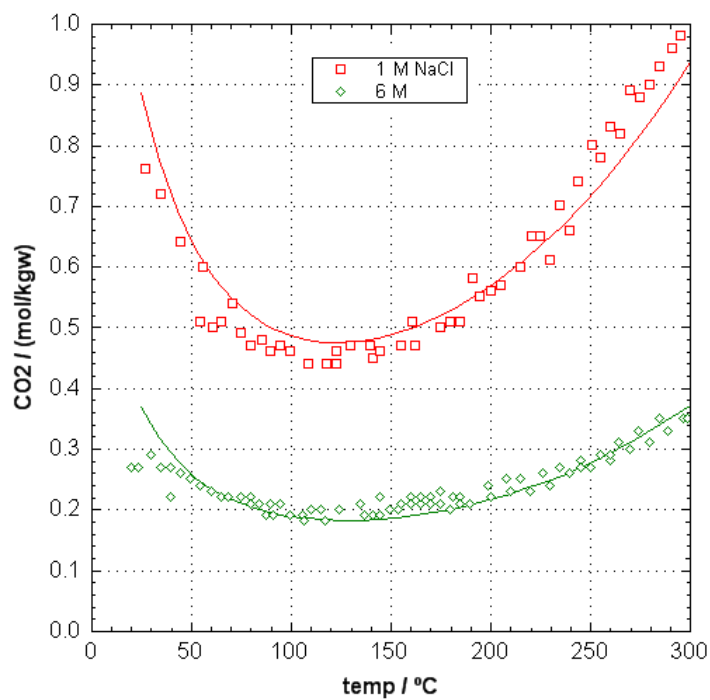


Figure A32. Solubility of CO₂ gas in 1 and 6 M NaCl solutions at about 40 atm CO₂. Measured data from Drummond, 1981. File Drummond.phr.

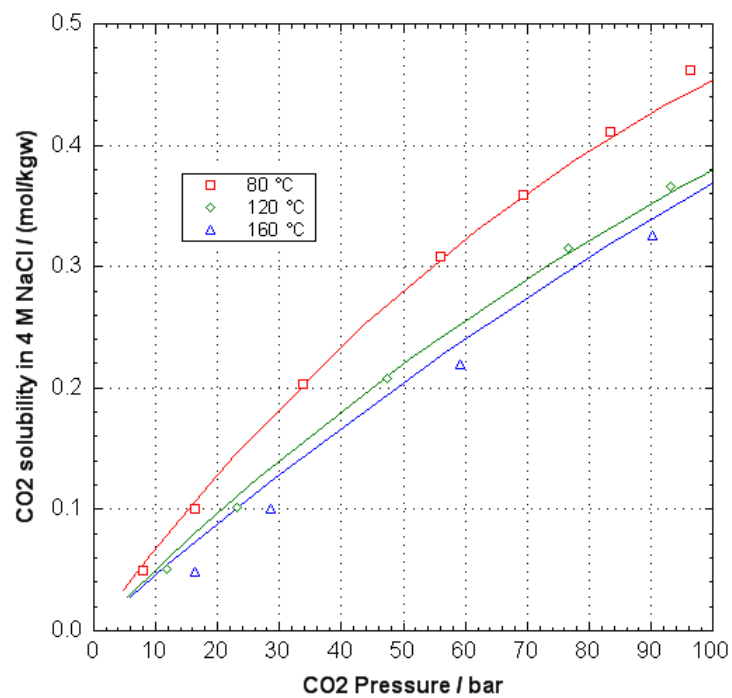


Figure A33. Solubility of CO₂ gas in 4 M NaCl solution. Measured data from Rumpf et al. 1994. File CO2_4M_NaCl.phr.

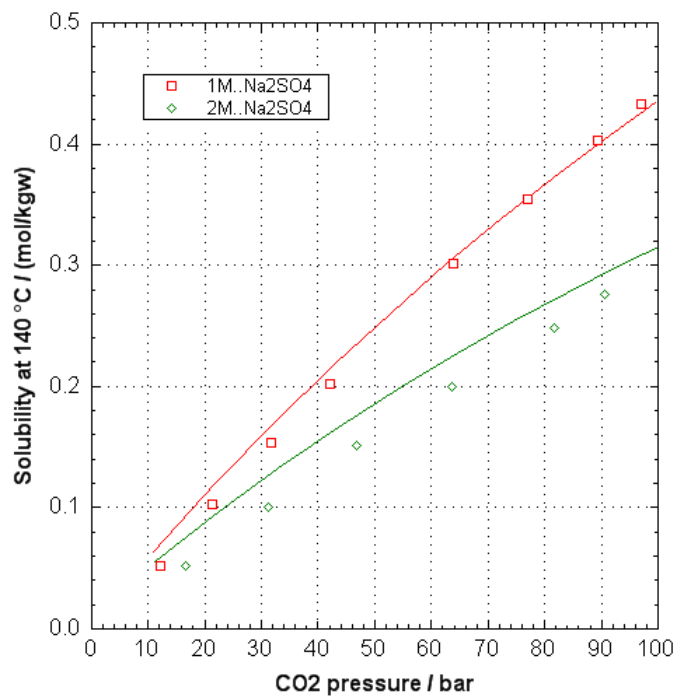


Figure A34. Solubility of CO₂ gas in Na₂SO₄ solutions at 140°C. Measured data from Rumpf and Maurer, 1993. File P_CO2_Na2SO4.phr.

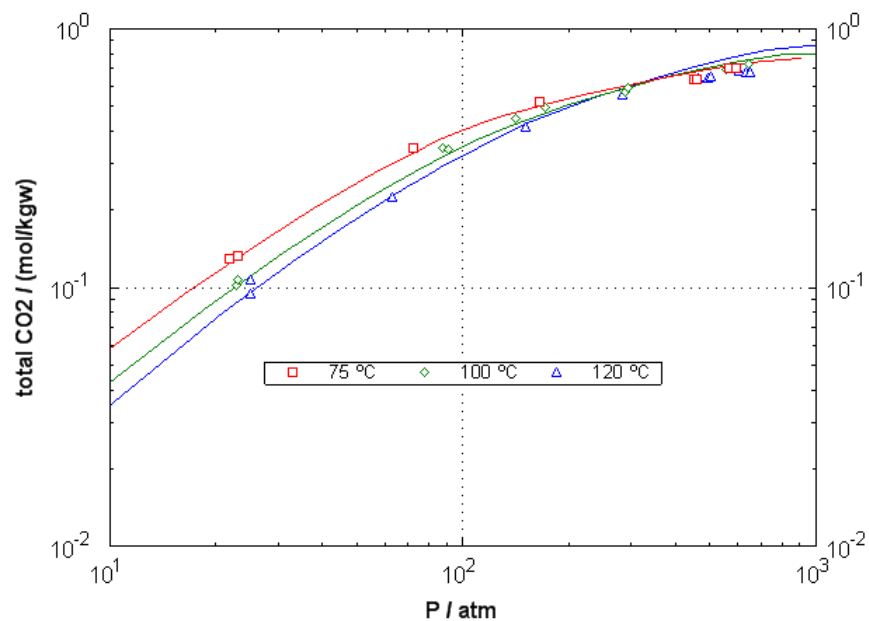


Figure A35. Solubility of CO₂ gas in 2.3 M CaCl₂ solution. Data from Springer et al., 2012. File CO2_CaCl2.phr.

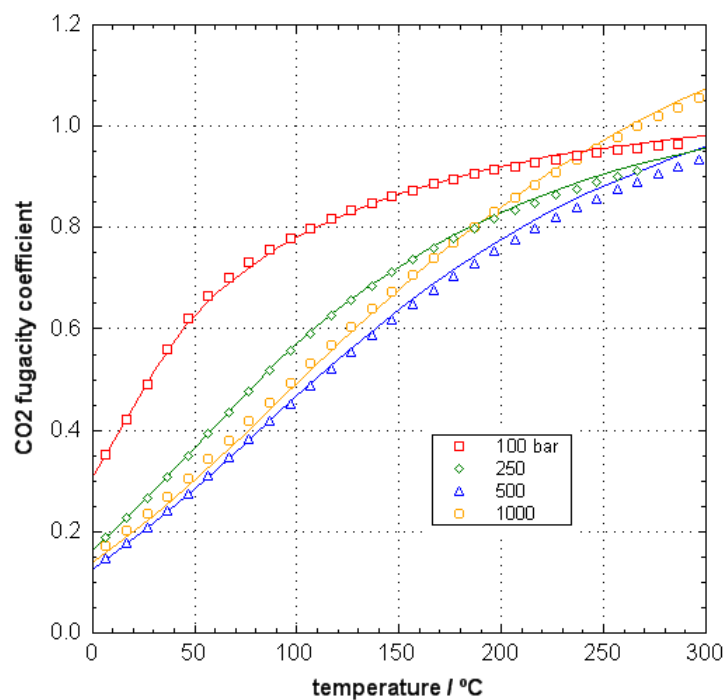


Figure A36. CO₂ fugacity coefficient as a function of temperature for CO₂ gas pressures from 100 - 1000 bar. Data from Angus et al., 1976. File phi_Angus_bar.phr.