

Experimental Data on Solubility of the Two Calcium Sulfates Gypsum and Anhydrite in Aqueous Solutions

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Abstract: Calcium sulfate exists in three forms, namely dihydrate or gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), anhydrite (CaSO_4), and hemihydrate or bassanite ($\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$) depending on temperature, pressure, pH, and formation conditions. The formation of calcium sulfates occurs widely in nature and in many engineering settings. Herein, a dataset containing the experimental solubility data of calcium sulfate minerals, i.e., gypsum and anhydrite, in aqueous solutions is presented. The compiled dataset contains calcium sulfates solubility values extracted from 42 papers published between 1906 and 2019. The dataset can be used for various scientific and engineering purposes such as environmental applications (e.g., gas treatment, wastewater treatment, and chemical disposal), geotechnical applications (e.g., clay-sulfate rock swelling), separation processes (e.g., crystallization, extractive distillation, and seawater desalination), and electrochemical processes (e.g., corrosion and electrolysis).



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1. Summary

The knowledge of the phase equilibria and solubility of inorganic salts in aqueous solutions is of interest to a wide range of scientific and technological disciplines. To date, many experimental studies have been conducted to measure solubility of calcium sulfates in pure water and electrolyte solutions. However, a dataset containing previously published experimental solubility values of calcium sulfates does not yet exist.

The dataset of the solubilities of calcium sulfates, i.e., gypsum and anhydrite, in aqueous solutions measured over a wide pressure–temperature–salt range were diligently and carefully compiled from the peer-reviewed literature. The gypsum and anhydrite solubility values were obtained through laboratory experiments that followed standard measurement methods. Thus, the compiled dataset can be used for various scientific and practical reasons.

Importance of the Dataset

- This dataset contains solubilities of gypsum and anhydrite under a broad range of conditions. The dataset can be used to tackle the calcium sulfate scaling potential for a variety of complex aqueous industrial streams.
- The dataset provides comprehensive insights about anhydrite dissolution and gypsum precipitation.
- The dataset is of interest for engineers and researchers in various disciplines such as chemistry, geochemistry, environmental and chemical engineering, geotechnical and civil engineering, etc.
- The data can be used to develop mathematical and machine learning models to determine calcium sulfate solubility in aquatic systems.

2. Literature Review

Cameron and Bell (1906) [1] studied the three-component system $\text{Ca}(\text{OH})_2\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ to determine solubility of gypsum at 25 °C. They prepared two types of solutions: (1) gypsum was added to lime solutions, and (2) lime was added to gypsum solutions. Partridge and White (1929) [2] measured solubilities of hemihydrate and anhydrite in water at temperatures ranging from 0 to 220 °C. The transition temperature of gypsum to anhydrite was reported to lie slightly below 40 °C. Campbell and Yanick (1932) [3] measured solubility of calcium sulphate in nickel sulphate solutions ($\text{NiSO}_4\text{-CaSO}_4\text{-H}_2\text{O}$ system) at 45, 75, and 90 °C. It was observed that at the same temperature calcium sulphate is more soluble in hot nickel sulphate solution than in water. Straub (1932) [4] measured solubility of calcium sulfate and calcium carbonate in water at temperatures between 182 and 316 °C. The author also investigated the influence of the presence of sodium sulfate on the solubility of calcium sulfate and calcium carbonate. It was observed that the solubility of calcium sulfate decreases with the increase in temperature.

Hill (1937) [5] determined solubility of anhydrite in water at 65, 45, and 35 °C with both supersaturation and undersaturation approaches. The transition point of anhydrite to gypsum was 42 °C. Hill and Wills (1938) [6] studied the system $\text{CaSO}_4\text{-Na}_2\text{SO}_4\text{-H}_2\text{O}$ at four temperatures; 25, 35, 50, and 75 °C. The authors obtained the solubility of gypsum, anhydrite, glauberite, the labile salt ($2\text{Na}_2\text{SO}_4\cdot\text{CaSO}_4\cdot 2\text{H}_2\text{O}$), and a hydrated sodium pentacalcium sulfate ($\text{Na}_2\text{SO}_4\cdot 5\text{CaSO}_4\cdot 3\text{H}_2\text{O}$). Booth and Bidwell (1950) [7] studied the solubility of six salts including calcium sulfate in water at high temperatures, between 141 and 408 °C [7]. Madgin and Swales (1956) [8] measured solubilities of gypsum and anhydrite in sodium chloride solutions ($\text{CaSO}_4\text{-NaCl-H}_2\text{O}$) with varied concentrations of up to saturation at 25 °C and for gypsum only at 35 °C.

Bock (1961) [9] determined solubilities of gypsum and anhydrite in concentrated aqueous solutions of NaCl at 25, 30, 40, and 50 °C. It was found that the transition temperature, which is 42 °C in pure water, decreases with increasing NaCl concentration. Dickson et al. (1963) [10] measured the solubility of anhydrite in H_2O from 96 to 263 °C and from 1 to 1000 bars. It was found that the solubility of anhydrite decreases with increasing the temperature and increases with increasing pressure. Marshall et al. (1964) [11] measured the solubilities of calcium sulfate and its hydrates in $\text{NaCl-H}_2\text{O}$ solutions at temperatures between 25 °C and 200 °C. Marshall and Slusher (1966) [12] measured the solubility of gypsum in various NaCl solutions at temperatures from 0 to 110 °C. Ostroff and Metler (1966) [13] measured solubility of gypsum in the system $\text{NaCl-MgCl}_2\text{-H}_2\text{O}$ at 28, 38, 50, 70, and 90 °C and at concentrations of up to 5.50 molal NaCl and 0.340 molal MgCl_2 in admixtures. Power and Fabuss (1966) [14] determined the change in solute concentration with time for 0.25 and 1.0 m NaCl in contact with gypsum, β -hemihydrate, β -soluble anhydrite, or insoluble anhydrite at 25 to 105 °C. Furby et al. (1968) [15] conducted experiments on the solubility of calcium sulfate in sodium chloride solutions and in simulated sea water solutions at temperatures of up to 100 °C.

Templeton and Rodgers (1967) [16] determined solubility of anhydrite in various salt solutions at temperatures and at pressures above the vapor pressure. The system $\text{CaSO}_4\text{-H}_2\text{O}$ was studied from 200 to 325 °C, and the system $\text{CaSO}_4\text{-NaCl-H}_2\text{O}$ was investigated at 250, 275, 300, and 325 °C from zero to about 6 molal NaCl. The other electrolytes were sodium nitrate, calcium chloride, magnesium chloride, sodium sulfate, and magnesium nitrate. Blount and Dickson (1969) [17] determined solubility of anhydrite in solutions with salt concentrations of up to 6.0 m NaCl at temperatures ranging from 73 to 451 °C and pressures up to 1410 bars. Block and Waters (1968) [18] acquired experimental solubility and phase data on the system $\text{CaSO}_4\text{-Na}_2\text{SO}_4\text{-NaCl-H}_2\text{O}$ at temperatures between 25 to 100 °C and at salinities of up to 4.0 m NaCl.

Tanji (1969) [19] measured solubility of gypsum in aqueous electrolyte systems (NaCl , MgCl_2 , CaCl_2 , Na_2SO_4 , MgSO_4 , CaSO_4 , or their mixtures) of limited ionic strength and at 25 °C. The author also developed a computer program to predict ion association and solubility of gypsum in simple and mixed aqueous electrolyte systems. Nakayama (1971) [20]

determined solubility of gypsum in NaClO_4 , NaCl , NaNO_3 , and NaOAc solutions and their mixtures through experimental and theoretical analysis.

Yeatts and Marshall (1972) [21] studied the solubility and association behavior of gypsum in the mixed electrolyte systems of Na_2SO_4 - NaCl - H_2O , Li_2SO_4 - LiNO_3 - H_2O , and Li_2SO_4 - LiCl - H_2O at 25 °C. Kalyanaraman et al. (1973a) [22] measured solubility of gypsum at 0.35 and 24.85 °C and solubility of anhydrite at 149.85, 249.85, and 349.85 °C in Na_2SO_4 + NaClO_4 + H_2O solutions at ionic strengths of up to 6 mol.kg⁻¹. Kalyanaraman et al. (1973b) [23] measured solubilities of gypsum at 24.85 °C and anhydrite at 149.85 and 249.85 °C in diluted and concentrated NaClO_4 - H_2O solutions having molalities from 0 to 6 mol.kg⁻¹. Blount (1973) [24] measured gypsum solubilities in H_2O solution at temperatures from 40 to 83 °C and pressures from 3 to 1004 bars.

Marshall and Slusher (1975) [25] measured solubilities of anhydrite in dilute and concentrated HNO_3 - H_2O solutions at temperatures from 100 to 350 °C. Innorta et al. (1980) [26] conducted solubility measurements to establish the gypsum–anhydrite equilibrium in the CaSO_4 - H_2O system at the atmospheric pressure. The saturation equilibrium was approached from both undersaturated and supersaturated solutions. Barba et al. (1984) [27] measured gypsum solubility in aqueous solutions containing Na_2SO_4 and MgCl_2 at 40 °C and the ionic strength of up to 7 m. Zhang and Muhammed (1989) [28] measured solubilities of gypsum in H_3PO_4 - HNO_3 , H_2SO_4 - HNO_3 , HNO_3 - $\text{Ca}(\text{NO}_3)_2$, H_3PO_4 - HNO_3 - $\text{Ca}(\text{NO}_3)_2$, and H_3PO_4 - HNO_3 - H_2SO_4 solutions at 25 °C [28]. Calmanovici et al. (1993) [29] measured solubilities of gypsum in mixtures of sulfuric and phosphoric acids at 20, 50, and 70 °C. Ling and Demopoulos (2004) [30] measured solubilities of calcium sulfate in H_2SO_4 solutions at a concentration range of 0 to 3.0 mol.L⁻¹ or (0 to 3.5 mol.kg⁻¹) at 100 °C.

Kumar et al. (2004) [31] measured different physicochemical properties such as solubilities, densities, viscosities, and electrical conductivities of gypsum for the system CaSO_4 - $2\text{H}_2\text{O}$ - CaCl_2 - H_2O at 30 °C. Kumar et al. (2005) [32] measured the thermodynamic, volumetric, transport, and surface properties, solubilities, densities, viscosities, electrical conductivities, and surface tensions of gypsum for the CaSO_4 - NaCl - H_2O system at 35 °C. Li and Demopoulos (2006) [33] investigated the effect of various chloride salts (NaCl , MgCl_2 , FeCl_2 , FeCl_3 , and AlCl_3) on the solubility of calcium sulfates in aqueous HCl or HCl - CaCl_2 solutions up to 79.85 °C. Kumar et al. (2007) [34] measured solubility of gypsum in diluted and concentrated NaCl solutions and constant CaCl_2 concentrations at 35 °C. Farrah et al. (2007) [35] measured solubility of gypsum on in acidic manganese sulfate solutions at temperatures from 30 to 105 °C.

Azimi and Papangelakis (2010) [36] measured the solubility of gypsum in electrolyte solutions containing NiSO_4 , H_2SO_4 , MgSO_4 , $\text{Fe}_2(\text{SO})_4$, LiCl , and HCl at 25, 45, 70, and 90 °C. Yuan et al. (2010) [37] measured solubilities of gypsum for the ternary systems of CaSO_4 - $2\text{H}_2\text{O}$ - NaOH - H_2O , CaSO_4 - $2\text{H}_2\text{O}$ - KOH - H_2O , and $\text{Ca}(\text{OH})_2$ - NaOH - H_2O over the temperature range of 25–75 °C. Wu et al. (2010) [38] measured solubility of gypsum in the system CaCl_2 - MgCl_2 - KCl and concentrations up to 3.86 m CaCl_2 , 1.17 m MgCl_2 , 0.27 m KCl , and their admixtures from 75 to 98 °C. Tian et al. (2010) [39] measured solubilities of gypsum in NH_4Cl , NH_4NO_3 , and mixed NH_4Cl and $(\text{NH}_4)_2\text{SO}_4$ solutions over the temperature range 25 to 70 °C. Wang et al. (2013) [40] measured the solubility isotherms of gypsum and insoluble anhydrite in the ternary system CaSO_4 - H_2SO_4 - H_2O at temperatures 24.95, 49.95, 74.95, and 89.95 °C. Shukla et al. (2018) [41] measured the solubility of gypsum in NaCl solutions in the presence of ammonium- or imidazolium-based ionic liquids (ILs), viz., ethylammonium lactate (EAL), 1-ethyl-3-methyl imidazolium hydrogen sulfate ($[\text{C}_2\text{mim}]\text{HSO}_4$), and 1-butyl-3-methyl imidazolium hydrogen sulfate ($[\text{C}_4\text{mim}]\text{HSO}_4$) at 30 °C. Scheuermann et al. (2019) [42] measured anhydrite solubility in low-density NaCl - H_2O solutions at 410 °C (26.5–32 MPa) and 450 °C (36.3–49.4 MPa).

3. Data Description

Herein, we conducted a systematic literature review on peer-reviewed experimental articles to compile a dataset including the experimental solubility of calcium sulfates in

pure water and various electrolyte solutions over wide ranges of pressure, temperature, and salt concentration. We presented a short description of the literature used to compile the dataset to make the dataset more useful for future applications. The dataset contains 4069 calcium sulfates solubility data extracted from 42 peer-reviewed papers published between 1906 and 2019.

The data on the pressure, temperature, and salt content of the aqueous solutions, namely H_2SO_4 , $\text{Ca}(\text{OH})_2$, NiSO_4 , CaSO_4 , Na_2SO_4 , NaCl , MgCl_2 , CaCl_2 , MgSO_4 , NaClO_4 , NaNO_3 , NaOAc , Li_2SO_4 , LiNO_3 , LiCl , NaClO_4 , HNO_3 , H_3PO_4 , $\text{Ca}(\text{NO}_3)_2$, FeCl_2 , FeCl_3 , AlCl_3 , HCl , $\text{Fe}_2(\text{SO})_4$, NaOH , KOH , KCl , NH_4Cl , NH_4NO_3 , $(\text{NH}_4)_2\text{SO}_4$, ammonium- or imidazolium-based ionic liquids (ILs), viz., ethylammonium lactate (EAL), 1-ethyl-3-methyl imidazolium hydrogen sulfate ($[\text{C}_2\text{mim}]\text{HSO}_4$), and 1-butyl-3-methyl imidazolium hydrogen sulfate ($[\text{C}_4\text{mim}]\text{HSO}_4$), obtained using standard laboratory testing methods, are provided.

Since this dataset references experimental studies spanning around 115 years, the solubility data have been expressed in many different units. It was not possible to provide the solubility values in one unit because, in most cases, the parameters (e.g., density) required for the conversion were not provided by the authors. Therefore, the solubility values are presented in their original units. Taherdangkoo et al. (2022) [43] provided a dataset containing 1912 experimental solubility data of the here-presented datasets but reported in a unified unit, namely molality. Their dataset includes calcium sulfate solubilities taken from [6,8,9,11–13,15,18,21,24,26,27,30–34,36–38,40,44,45].

The solubility data are divided into 42 groups, with each having data collected from the same paper. The tables shown within each group are representing the tables in the corresponding paper. This would allow for an easy assessment and analysis of calcium sulfate solubilities obtained from various sources. The dataset for each group is saved in individual tabs within the .xlsx file, and the tab names (notations) correspond to the descriptions presented in Table 1. Note that the groups are listed in the .xlsx file and Table 1 according to the date of publication. Table 1 also provides a detailed description of each group within the dataset.

Table 1. Description of groups of datasets.

Dataset	Solutions	T	Calcium Sulfate (s)	Time	Source
DS-1	Ca(OH) ₂	25 °C	DH	two weeks	Cameron and Bell (1906) [1]
DS-2	Water	98.6–198.4 °C	AH	1.5–24 h	Partridge and White (1929) [2]
DS-3	NiSO ₄	45, 75, 90 °C	DH	24 h	Campbell and Yanick (1932) [3]
DS-4	Water	182–316 °C	DH	90 h	Straub (1932) [4]
DS-5	K ₂ SO ₄	35, 45, 65 °C	AH	4 months	Hill(1937) [5]
DS-6	Na ₂ SO ₄	25, 35, 50, 70 °C	DH	12 h	Hill and Wills (1938) [6]
DS-7	Water	0–408 °C	AH	20 h	Booth and Bidwell (1950) [7]
DS-8	NaCl, Na ₂ SO ₄	25, 35 °C	DH + AH	less than 60 h	Madgin and Swales (1956) [8]
DS-9	NaCl	25, 30, 40, 50 °C	DH + AH	96 h	Bock(1961) [9]
DS-10	Water	100–275 °C	AH	17–500 h	Dickson et al. (1963) [10]
DS-11	NaCl	40–200 °C	DH + AH	1 h–5 days	Marshall et al. (1964) [11]
DS-12	NaCl	0–110 °C	DH	16–96 h	Marshall and Slusher (1966) [12]
DS-13	MgCl ₂ , NaCl	28–70 °C	DH	9 days	Ostroff and Metler (1966) [13]
DS-14	NaCl	25–105 °C	DH + AH	0.5–246 h	Power and Fabuss (1966) [14]
DS-15	Sea salt, NaCl	25, 75, 100 °C	DH + AH	—	Furby et al. (1968) [15]
DS-16	NaCl, CaCl ₂ , Na ₂ SO ₄ , MgSO ₄ , MgCl ₂ , NaNO ₃ , or their mixtures	250, 275, 300, 325 °C	AH	—	Templeton and Rodgers (1967) [16]
DS-17	NaCl	100–450 °C	AH	24–1080 h	Blount and Dickson (1969) [17]
DS-18	NaCl, Na ₂ SO ₄	25–100 °C	DH	at least 3 weeks	Block and Waters (1968) [18]
DS-19	NaCl, MgCl, CaCl ₂ , Na ₂ SO ₄ , MgSO ₄ , CaSO ₄ , or their mixtures	25 °C	DH	24 h	Tanji (1969) [19]
DS-20	NaClO ₄ , NaCl, NaNO ₃ , NaOAc	25 °C	DH	at least 1 week	Nakayama (1971) [20]
DS-21	Na ₂ SO ₄ , NaCl, Li ₂ SO ₄ , LiNO ₃ , LiCl, NaNO ₃	25 °C	DH	18–112 h	Yeatts and Marshall (1972) [21]
DS-22	Na ₂ SO ₄ , NaClO ₄	273–623 K	DH + AH	—	Kalyanaraman et al. (1973a) [22]
DS-23	NaClO ₄	298–523 K	DH + AH	—	Kalyanaraman et al. (1973b) [23]

Table 1. Cont.

Dataset	Solutions	T	Calcium Sulfate (s)	Time	Source
DS-24	Water	50–83 °C	DH + AH	—	Blount (1973) [24]
DS-25	HNO ₃	100–350 °C	DH + AH	3–23 h	Marshall and Slusher (1975) [25]
DS-26	Water	25–65 °C	DH + AH	one week	Innorta et al. (1980) [26]
DS-27	MgCl ₂ , Na ₂ SO ₄	40 °C	DH	36 h	Barba et al. (1984) [27]
DS-28	H ₃ PO ₄ , HNO ₃ , H ₂ SO ₄ , Ca(NO ₃) ₂	25 °C	DH	3–4 weeks	Zhang and Muhammed (1989) [28]
DS-29	H ₃ PO ₄ , H ₂ SO ₄	20, 50, 70 °C	DH	12 h	Calmanovici et al. (1993) [29]
DS-30	H ₂ SO ₄	100 °C	DH + AH	2 h	Ling and Demopoulos (2004) [30]
DS-31	CaCl ₂	30 °C	DH	24 h	Kumar et al. (2004) [31]
DS-32	NaCl	35 °C	DH	24 h	Kumar et al. (2005) [32]
DS-33	NaCl, MgCl ₂ , AlCl ₃ , FeCl ₂ , FeCl ₃ , AlCl ₃ , HCl	298.1–353 K	DH + AH	1–5 h	Li and Demopoulos (2006) [33]
DS-34	CaCl ₂ +NaCl	35 °C	DH		Kumar et al. (2007) [34]
DS-35	Mn ²⁺ , H ₂ SO ₄	30–105 °C	DH + AH	4–36 days	Farrah et al. (2007) [35]
DS-36	NiSO ₄ , H ₂ SO ₄ , MgSO ₄ , Fe ₂ (SO ₄) ₃ , LiCl and HCl	25–90 °C	DH + AH	24 h	Azimi and Papangelakis (2010) [36]
DS-37	NaOH, KOH	25–75 °C	DH	6.5 h	Yuan et al. (2010) [37]
DS-38	CaCl ₂ , MgCl ₂ , KCl	348.15–371.15 K	DH	6 h	Wu et al. (2010) [38]
DS-39	NH ₄ Cl, NH ₄ NO ₃ , mixed NH ₄ Cl and (NH ₄) ₂ SO ₄	298.15–343.15 K	DH	24 h	Tian et al. (2012) [39]
DS-40	H ₂ SO ₄	298.1–363.1 K	DH + AH	128 h	Wang et al. (2013) [40]
DS-41	NaCl+ethylammonium lactate (EAL)/1-ethyl-3-methylimidazolium hydrogen sulfate ([C ₂ mim]HSO ₄)/1-butyl-3-methylimidazolium hydrogen sulfate ([C ₄ mim]HSO ₄)	30 °C	DH	—	Shukla et al. (2018) [41]
DS-42	NaCl	410, 450 °C	AH	—	Scheuermann et al. (2019) [42]

4. Methods

A systematic literature review was conducted to obtain peer-reviewed papers on experimental studies of calcium sulfate solubilities in various aquatic systems. The data were collected from the studies that employed the standard testing techniques and followed recommended procedures to measure the solubility of calcium sulfates in aqueous solutions. The methodology of data collection involved screening articles' titles, keywords, and abstracts that met the criteria of experimental calcium sulfate solubilities. All the data within the dataset were derived from tables. The data are presented in the .xlsx in similar formats reported in the articles. We did not include research articles that provided solubility values through graphs to avoid introducing any errors that may occur during the data extraction process.

5. User Notes

The data presented in the dataset are collated from tables published in various references, which can be freely and easily used by interested readers.

Author Contributions: R.T.: Conceptualization, Methodology, Data curation, Validation, Supervision Writing—original draft; M.T.: Data curation, Validation, Writing—review and editing; A.S.: Data curation, Writing—review and editing, T.M.: Writing—review and editing, H.Y.: Validation, Writing—review and editing, C.B.: Funding acquisition, Supervision, Writing—review and editing. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The experimental data are available at Mendeley Data repository with doi: 10.17632/phmrfngcxn.1. The direct URL to data: <https://data.mendeley.com/datasets/phmrfngcxn/1> (accessed on 22 September 2022).

Conflicts of Interest: The authors declare that they have no known competing financial interest or personal relationships that could have appeared to influence the work reported in this paper.

Ethics Statements: The authors declare that the present work did not include experiments on human subjects and/or animals.

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