CARBOSILICATES

stoichiometry, CO_2 content, crystal structure and thermal stability

A. Cuneyt Tas, *Ph.D.*

http://www.cuneyttas.com

https://www.linkedin.com/in/a-cuneyt-tas-8a971118

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Underlined blue texts throughout have hyperlinks.

Carboaluminate phases of hydrated Portland cement that contains calcium carbonate in it are actually carbonate-containing (*i.e.*, the CO_3^{2-} anions incorporated in the solid crystals of the newly formed phases) calcium aluminate compounds. These phases are formed especially by the reaction of the hydration product(s) of the C_3A (tricalcium aluminate) component of Portland cement with the free calcium carbonate particles/grains present in the cement.

Although the calcium **carboaluminate** phases [1, 2] are well-known, the literature seemed to remain shy of naming the below four compounds

scawtite,
spurrite,
galuskinite
and
tilleyite as calcium carbosilicates.

- This note names, for the first time in the open digital or paper literature, the compounds scawtite, spurrite, galuskinite and tilleyite as calcium carbosilicates or simply **carbosilicates**.

[1] A. Ipavec, R. Gabrovsek, T. Vuk, V. Kaucic, J. Macek, and A. Meden, "Carboaluminate phases formation during the hydration of calcite-containing Portland cement," *J. Am. Ceram. Soc.*, 94, 1238-1242 (2011).
[2] F. Georget, B. Lothenbach, W. Wilson, F. Zunino, and K. Scrivener, "Stability of hemicarbonate under cement paste-like conditions," *Cement Concrete Res.*, 153, 106692 (2022).

Carboaluminates (as encountered during the hydration of the C_3A of Portland cement containing calcium carbonate) are [1, 2]:

(1) hemicarboaluminate ($Ca_4Al_2(OH)_{13}(CO_3)_{0.5}\cdot 5.5H_2O$)
(2) monocarboaluminate ($Ca_4Al_2O_6(CO_3)\cdot 11H_2O$).

The above couple belongs to the crystallographic group of the naturally-occurring mineral hydrocalumite ($Ca_4Al_2(OH)_{12}(Cl, CO_3, OH)_2\cdot 4H_2O$). Hydrotalcite ($Mg_6Al_2(CO_3)(OH)_{16}\cdot 4H_2O$).

- There are four well-characterized [3, 4] and naturally-occurring **carbosilicates**:

(1) scawtite ($Ca_7(Si_3O_9)CO_3\cdot 2H_2O$) → belongs to the $CaO\text{-}SiO_2\text{-}CO_2\text{-}H_2O$ quaternary system
(2) tilleyite ($Ca_5(Si_2O_7)(CO_3)_2$)
(3) spurrite ($Ca_5(SiO_4)_2CO_3$)
(4) galuskinite ($Ca_7(SiO_4)_3CO_3$).

} belong to the $CaO\text{-}SiO_2\text{-}CO_2$ ternary system

[1] A. Ipavec, R. Gabrovsek, T. Vuk, V. Kaucic, J. Macek, and A. Meden, "Carboaluminate phases formation during the hydration of calcite-containing Portland cement," *J. Am. Ceram. Soc.*, 94, 1238-1242 (2011).
[2] F. Georget, B. Lothenbach, W. Wilson, F. Zunino, and K. Scrivener, "Stability of hemicarbonate under cement paste-like conditions," *Cement Concrete Res.*, 153, 106692 (2022).
[3] J. D. Grice, "The structure of spurrite, tilleyite and scawtite, and relationships to other silicate-carbonate minerals," *Can. Mineral.*, 43, 1489-1500 (2005).
[4] B. Lazic, T. Armbruster, V. B. Svelyeva, A. E. Zadov, N. N. Pertsev, and P. Dzierzanowski, "Galuskinite, $Ca_7(SiO_4)_3CO_3$, a new skarn mineral from the Birkhin gabbro massif, Eastern Siberia, Russia," *Mineral. Magazine*, 75, 2631-2648 (2011).

There are a few US patents or patent applications [5-7] that describe high temperature-high pressure processes to synthesize some of the carbosilicates of the previous page.

[5] V. Atakan, “Steam-assisted production of metal silicate cements, compositions and methods thereof,” U.S. Patent No: 10,752,545 B2, August 25, 2020
[6] R. E. Riman and D. Kopp, “Methods for low energy inorganic material synthesis,” U.S. Patent No: 11,332,847 B2, May 17, 2022
[7] R. E. Riman and D. Kopp, “Methods for low energy inorganic material synthesis,” U.S. Patent application number: 2023/0115706 A1, April 13, 2023

Comparison of the stoichiometry of carbosilicates

(with those of tobermorite and wollastonite)

Spurrite: $Ca_5(SiO_4)_2CO_3$ or $5CaO{\cdot}2SiO_2{\cdot}CO_2$ — *neso*carbosilicate
63.07 wt% CaO – 27.03% SiO_2 – **9.90**% CO_2
Ca-to-Si molar ratio: 2.500

Galuskinite: $Ca_7(SiO_4)_3CO_3$ or $7CaO{\cdot}3SiO_2{\cdot}CO_2$ — *neso*carbosilicate
70.51 wt% CaO – 32.38% SiO_2 – **7.91**% CO_2
Ca-to-Si molar ratio: 2.333

Tilleyite: $Ca_5(Si_2O_7)(CO_3)_2$ or $5CaO{\cdot}2SiO_2{\cdot}2CO_2$ — *soro*carbosilicate
57.39 wt% CaO – 24.60% SiO_2 – **18.02**% CO_2
Ca-to-Si molar ratio: 2.500

Scawtite: $Ca_7(Si_3O_9)_2CO_3{\cdot}2H_2O$ or $7CaO{\cdot}6SiO_2{\cdot}CO_2{\cdot}2H_2O$ — *cyclo*carbosilicate
47.12 wt% CaO – 43.27% SiO_2 – **5.28**% CO_2 – 4.32% H_2O
Ca-to-Si molar ratio: 1.167

Wollastonite-2M: $CaSiO_3$ or $Ca_3(Si_3O_9)$ or $CaO{\cdot}SiO_2$ or $3CaO{\cdot}3SiO_2$ — *cyclo*silicate
48.28wt% CaO – 51.72% SiO_2
Ca-to-Si molar ratio: 1.000

Tobermorite-11 Å [7]: $Ca_{4.5}Si_6O_{16}(OH){\cdot}5H_2O$ — *ino*silicate
35.45 wt% CaO – 50.64% SiO_2
Ca-to-Si molar ratio: 0.750

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[8] S. Merlino, E. Bonaccorsi and T. Armbruster, “The real structure of tobermorite 11 Å: normal and anomalous forms, OD character and polytypic modifications,” *Eur. J. Mineral.*, 13, 577-590 (2001).

It is important to note that the formation of anhydrous carbosilicates galuskinite, spurrite and tilleyite requires significant amounts of CO_2 (if and only if it is taken from the gaseous atmosphere) to be available at the nucleation front and then it must be chemically accommodated as carbonate (CO_3^{2-}) anions in the solidified crystal structures of carbosilicates. Such a chemical accommodation does not resemble at all to the physical absorption/attachment (*via* van der Waals forces) of CO_2 molecules onto the active, surface sites of any material.

Tilleyite:	18.02 wt% CO_2
Spurrite:	9.90 wt% CO_2
Galuskinite:	7.91 wt% CO_2

On the other hand, **scawtite** (which is a hydrated carbosilicate) has a theoretical (maximum) CO_2 content of only **5.28%** in its crystal structure, which is a lower figure than those of the anhydrous carbosilicates.

Crystal structures and lattice parameters of three carbosilicates scawtite, spurrite, tilleyite, (and of wollastonite-2M and tobermorite-11 Å)

SCAWTITE
Monoclinic
a=6.631 Å
b=15.195 Å
c=10.121 Å
α=90°
β=100.59°
γ=90°
V=1002.403 Å^3
Density=2.760 g/cm^3
Space group: I 1 m1
AMCSD No: 0006058

SPURRITE
Monoclinic
a=10.490 Å
b=6.705 Å
c=14.160 Å
α=90°
β=101.317°
γ=90°
V=976.585 Å^3
Density=3.024 g/cm^3
Space group: P1 21/a1
AMCSD No: 0009239

TILLEYITE
Monoclinic
a=15.082 Å
b=10.236 Å
c=7.572 Å
α=90°
β=105.170°
γ=90°
V=1128.226 Å^3
Density=2.876 g/cm^3
Space group: P1 21/a1
AMCSD No: 0006060

WOLLASTONITE-2M
Monoclinic
a=15.424 Å
b=7.324 Å
c=7.069 Å
α=90°
β=95.371°
γ=90°
V=795.069 Å^3
Density=2.911 g/cm^3
Space group: P1 21/a1
AMCSD No: 0007360

TOBERMORITE-11 Å
Monoclinic
a=6.735 Å
b=7.385 Å
c=22.487 Å
α=90°
β=90°
γ=123.350°
V=935.351 Å^3
Density=2.460 g/cm^3
Space group: B 1 1 m
AMCSD No: 0002330

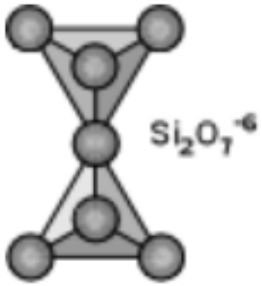
GALUSKINITE
Monoclinic
a=18.787 Å
b=6.724 Å
c=10.467 Å
α=90°
β=90.788°
γ=90°
V=1322.237 Å^3
Density=3.108 g/cm^3
Space group: P1 21/c1
AMCSD No: 0018535

- AMCSD stands for the "American Mineralogist Crystal Structure Database." Upon typing the name of mineral at this website, the search will generate a list of the CIF data (crystallographic information files) available. Each and every CIF file has a unique number such as those shown here.

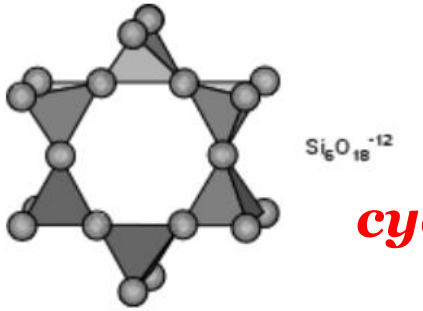
- Lattice parameters given as a, b, c state the dimensions of the unit cells (*crystallographic*) along the x, y, z Cartesian coordinates.

What are *neso*-, *soro*-, and *cyclo*-silicates?

neso The corner oxygens of a silicate tetrahedron are not shared with other silicate tetrahedra present in the structure. Each tetrahedron is isolated. The compound's formula contains SiO_4 groups.



soro Only one of the corner oxygens of silicate tetrahedra are shared with another silicate tetrahedron. present in the structure. The compound's formula contains Si_2O_7 groups. Such are also called "pyro-silicates."



cyclo Two of the corner oxygens of every silicate tetrahedron are shared with another silicate tetrahedron and the structure is arranged as a ring. The compound's formula contains Si_3O_9 (or Si_6O_{18} or Si_5O_{15}) groups.

- Spurrite and galuskinite are *neso*silicates. While tilleyite is a sorosilicate, scawtite is a *cyclo*silicate just like wollastonite. The sketches are from the below reference.

[9] S. A. Nelson, "Silicate structures, structural formula; neso-, cyclo-, and soro-silicates," Tulane University, New Orleans, Louisiana, USA
https://www2.tulane.edu/%7Esanelson/eens211/silicate_structures08.htm

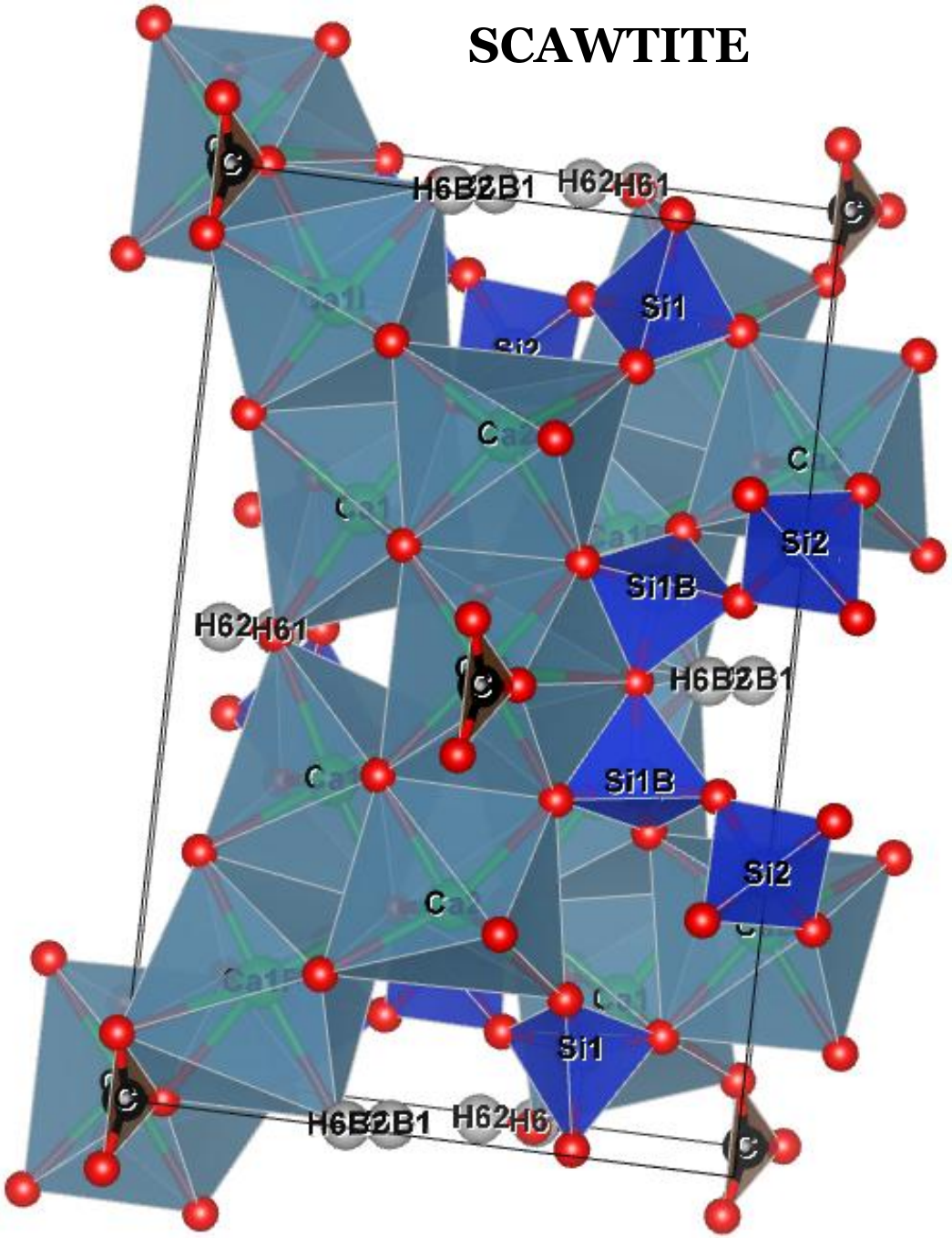
The crystal structure drawings of the six compounds cited above are provided in the following slides.

CIF (crystallographic information file)-based structure information is first obtained from the open access AMCSD (American Mineralogist Crystal Structure) database and then the respective CIF files (*e.g.*, the CIF file numbered as 0002330 for tobermorite-11 Å summarized two pages before) were opened in the shareware software VESTA [10] to plot the structures and to obtain the XRD patterns, *hkl*, 2θ and peak intensity values.

The crystallographic unit cells given in the next slides are the "repeating motifs" of the crystal of a given material. Continuing with the example of tobermorite-11 Å (which is the major compound of very high porosity (75-80% porosity) industrial autoclaved aerated concrete (AAC) responsible from its strength), as one notices from the AMCSD CIF file 0002330, the tobermorite unit cell has a height (lattice parameter c) of 22.487 Å, which is equal to 2.2487×10^{-6} millimeter. This means that in a 1 millimeter-long pure tobermorite single crystal there will be 444,701 (444.7 thousand) of those repeating motif unit cells that are tightly stacked with one another. This is how these drawings we will present soon are tied to the reality. Most of the chemical and physical properties of materials stem from their crystallographic unit cells.

[10] K. Momma and F. Izumi, "VESTA 3 for three dimensional visualization of crystal, volumetric and morphology data," *J. Appl. Crystallogr.*, 44, 1272-1276 (2011).

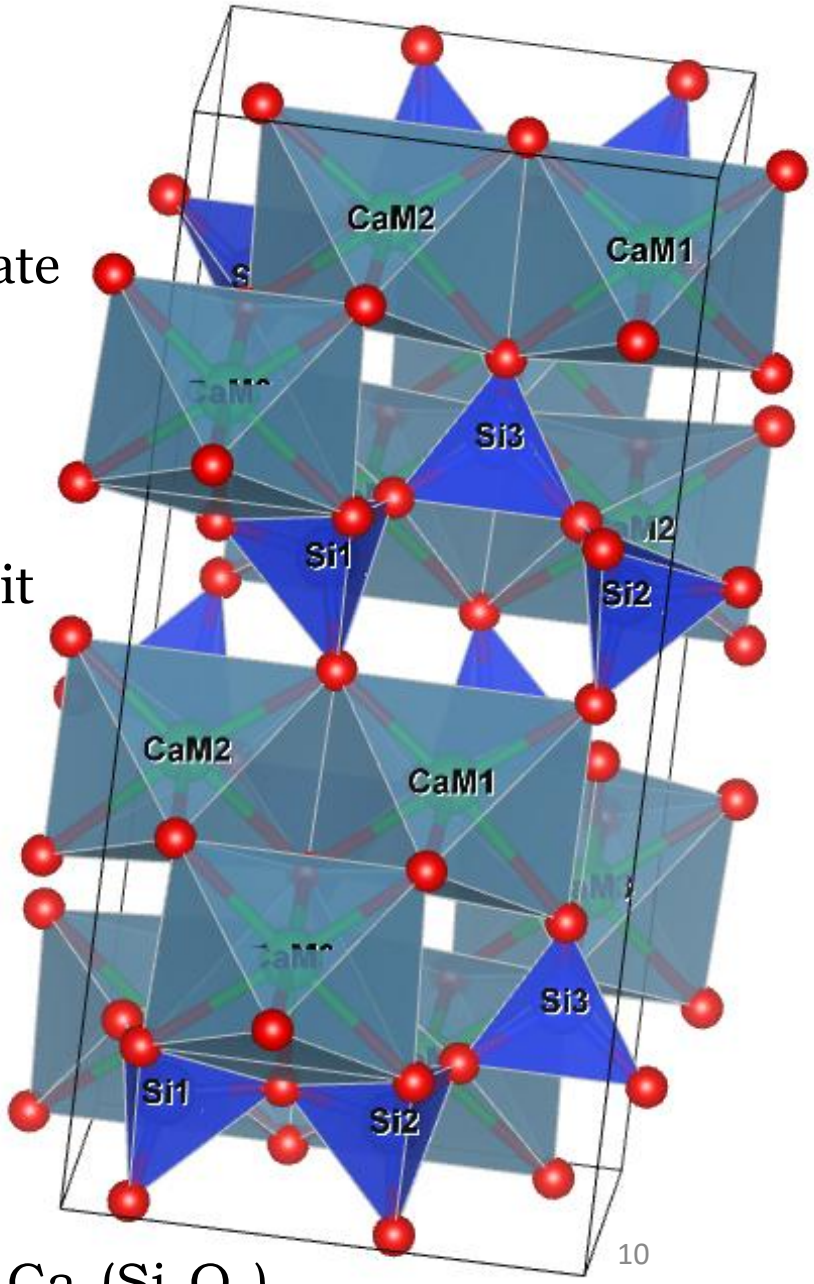
SCAWTITE



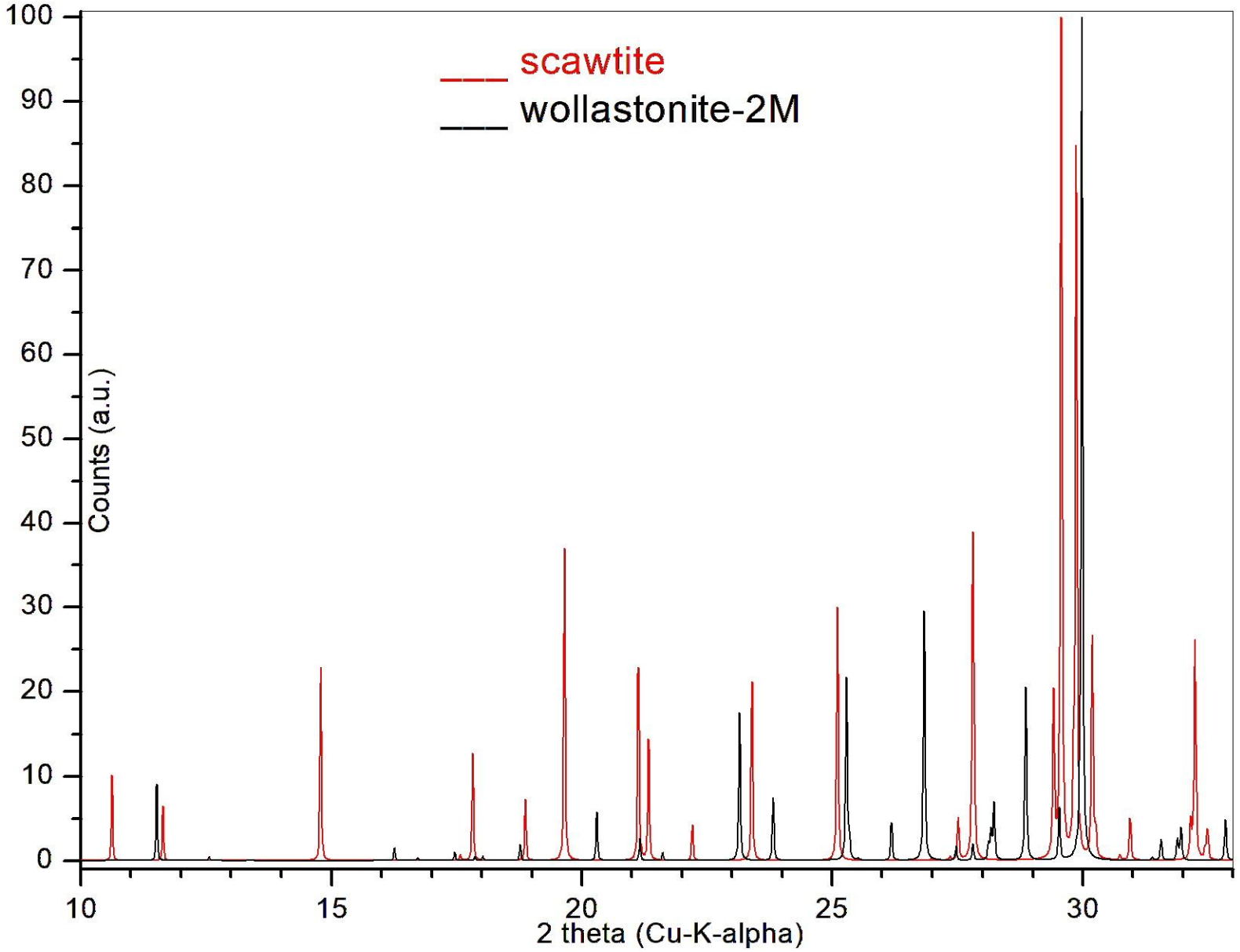
$Ca_7(Si_3O_9)_2CO_3 \cdot 2H_2O$

Both of scawtite and wollastonite–2M are ***cyclo*silicates**. The SiO_4 tetrahedra in blue form **rings** by corner sharing. CO_3^{2-} groups of carbosilicate scawtite are shown as triangles (black: carbon, red: oxygens). Waters (grey) and CO_3^{2-} groups are located close to the unit cell faces or corners, which typically results in **thermodynamically easier** dehydration-rehydration and decarbonation-carbonation events.

WOLLASTONITE – 2M



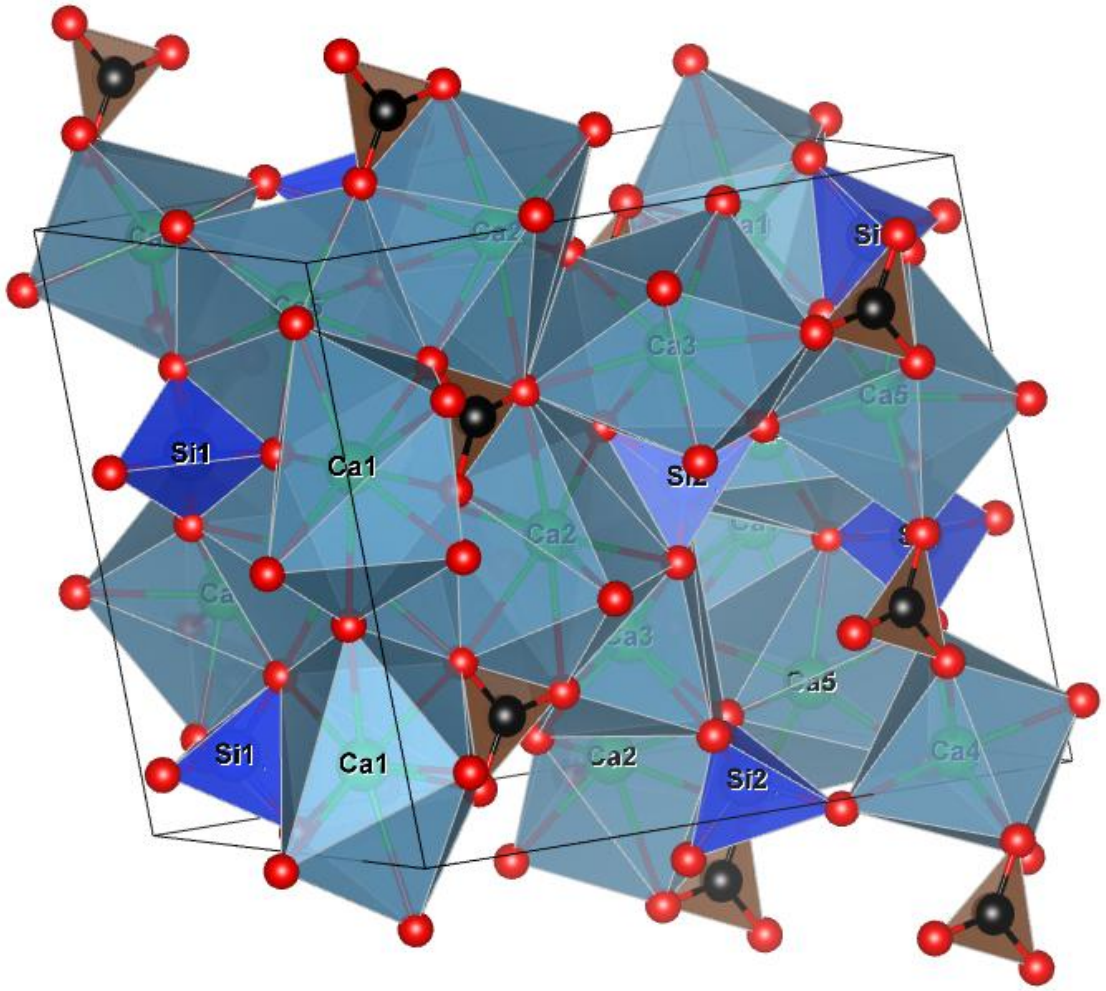
$Ca_3(Si_3O_9)$



Scawtite			Wollastonite-2M		
h k l	2θ	I/I$_0$	*h k l*	2θ	I/I$_0$
(1 1 0)	14.777	17	(2 0 0)	11.516	7
(0 0 -2)	17.817	10	(1 1 1)	18.768	2
(0 3 1)	19.652	30	(2 1 -1)	20.298	5
(0 3 -1)	19.652	30	(4 0 0)	23.149	15
(0 4 0)	23.399	36	(2 0 -2)	26.841	28
(1 1 2)	25.107	27	(1 1 -2)	28.221	7
(2 1 -1)	27.799	27	(2 0 2)	28.858	20
(1 4 1)	29.412	19	(2 1 -2)	29.525	6
(0 4 2)	29.564	99	(3 2 0)	29.978	100
(0 4 -2)	29.564	100	(5 1 0)	31.563	3
(2 0 -2)	29.863	83	(3 1 -2)	31.887	3
(-1 3 -2)	30.186	27	(3 2 -1)	31.959	4
(0 3 -3)	32.239	27	(4 0 -2)	32.849	5

The XRD patterns (obtained using Vesta) of scawtite (of its structure and CIF given in AMCSD 0006058) and wollastonite-2M (based on the CIF of AMCSD 0007360) are plotted here. The table provides the most intense X-ray peaks, that will be observed for each phase, over the 2θ range of 10 to 33°. This 2θ range of 10 to 33° is enough to identify the individual phases of scawtite and wollastonite if they were in a hypothetical two phase mixture.

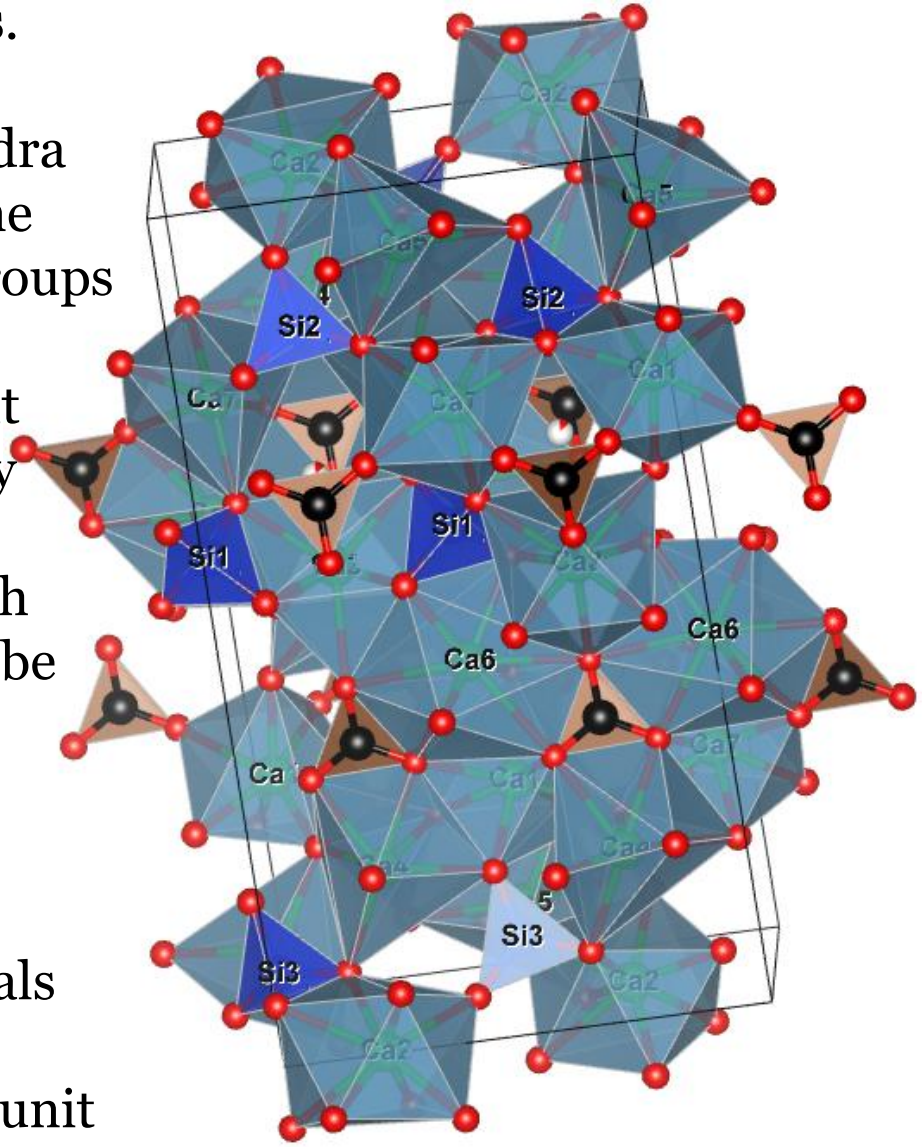
SPURRITE



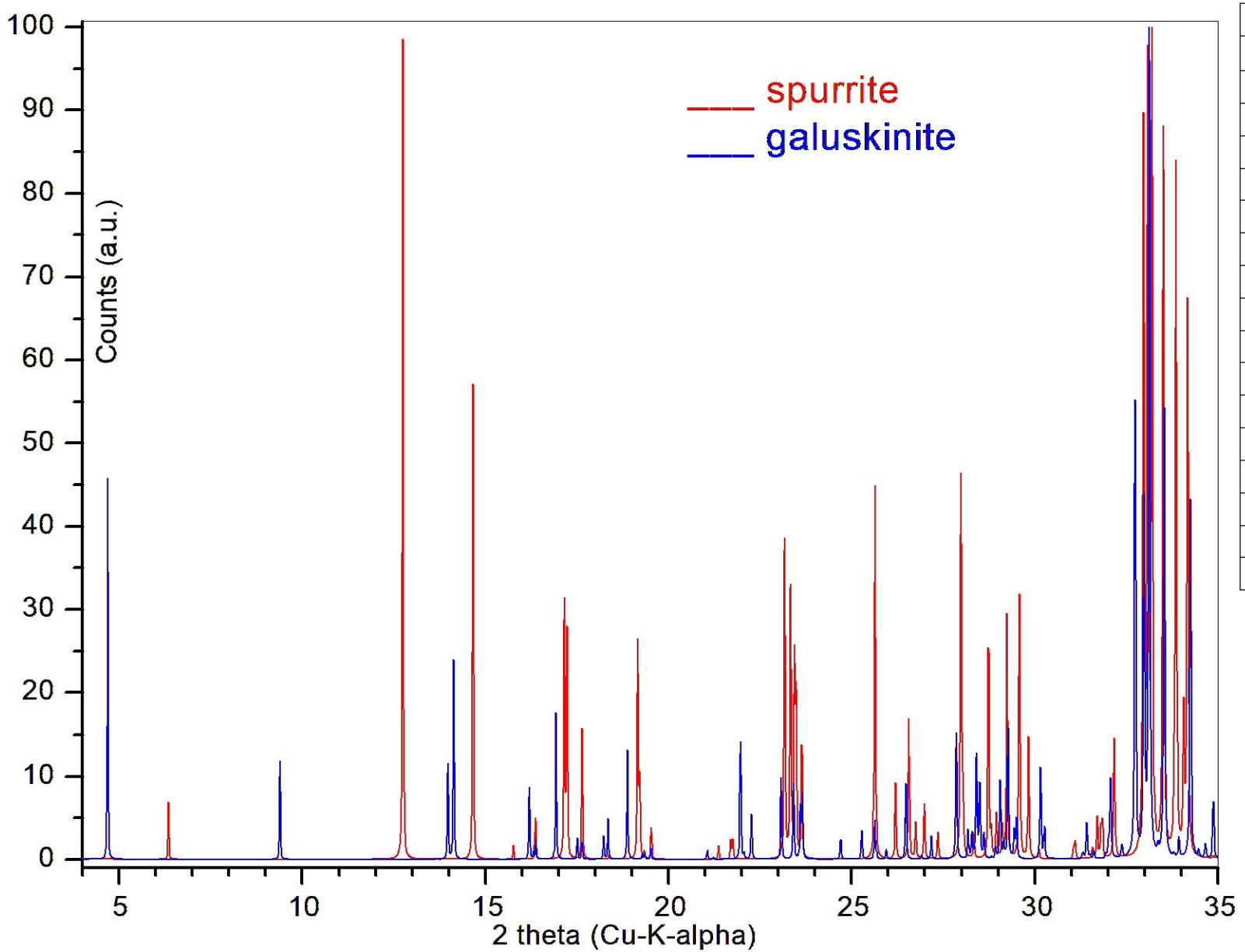
$Ca_5(SiO_4)_2CO_3$

Both of spurrite and galuskinite are ***neso*carbosilicates**. None of the blue SiO_4 tetrahedra share corners with one another. The CO_3^{2-} groups of both minerals are located inside the unit cells and this typically causes the Decarbonation of such minerals to occur (to be observed by TGA or combined TGA-FTIR analysis) at higher temperatures (in comparison to minerals whose CO_3^{2-} groups are located along the unit cell faces or corners).

GALUSKINITE



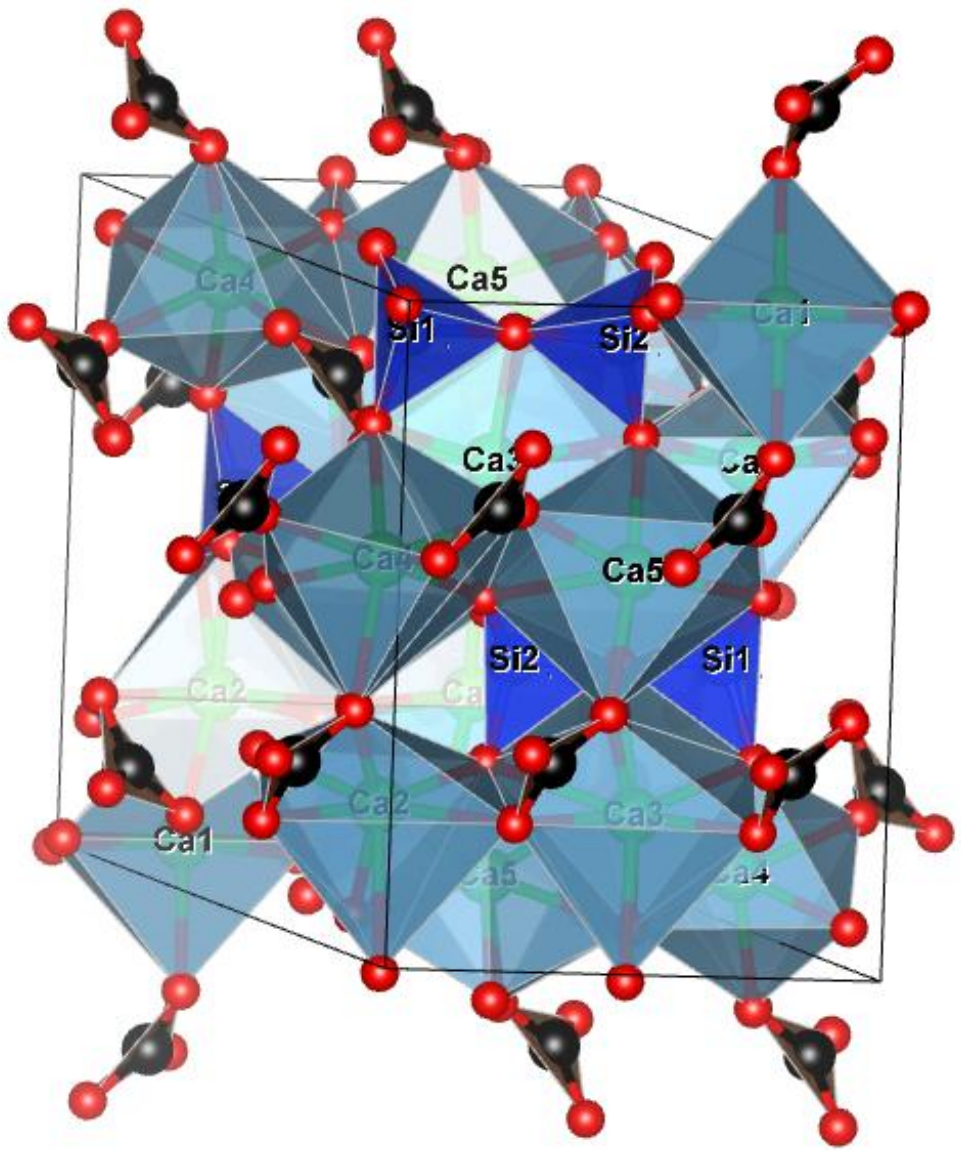
$Ca_7(SiO_4)_3CO_3$



Spurrite			Galuskinite		
h k l	2θ	I/I₀	*h k l*	2θ	I/I₀
(0 0 1)	6.361	5	(1 0 0)	4.700	51
(0 0 2)	12.741	70	(3 0 0)	14.132	31
(0 1 1)	14.659	42	(0 0 2)	16.929	24
(2 0 -1)	17.157	24	(4 0 0)	18.881	18
(0 0 3)	19.161	21	(3 0 -2)	21.967	20
(2 0 -3)	23.176	34	(4 1 0)	23.081	15
(0 1 3)	23.335	29	(0 2 0)	26.489	15
(2 0 2)	23.441	20	(0 2 1)	27.848	25
(0 0 4)	25.643	41	(5 1 -1)	28.396	21
(1 2 0)	27.970	43	(1 1 3)	29.262	27
(1 1 -4)	28.724	25	(3 1 -3)	32.061	17
(0 2 2)	29.566	32	(4 2 0)	32.731	98
(2 2 -2)	33.074	97	(2 2 -2)	32.976	78
(2 2 1)	33.189	100	(6 0 -2)	33.113	100
(2 0 4)	33.844	90	(6 0 2)	33.528	99
(4 0 -1)	34.165	73	(0 0 4)	34.242	81

The XRD patterns (obtained using Vesta) of spurrite (of its structure and CIF given in AMCSD 0009239) and galuskinite (based on the CIF of AMCSD 0018535) are plotted here. The table provides the most intense X-ray peaks, that will be observed for each phase, over the 2θ range of 4 to 35°.

TILLEYITE



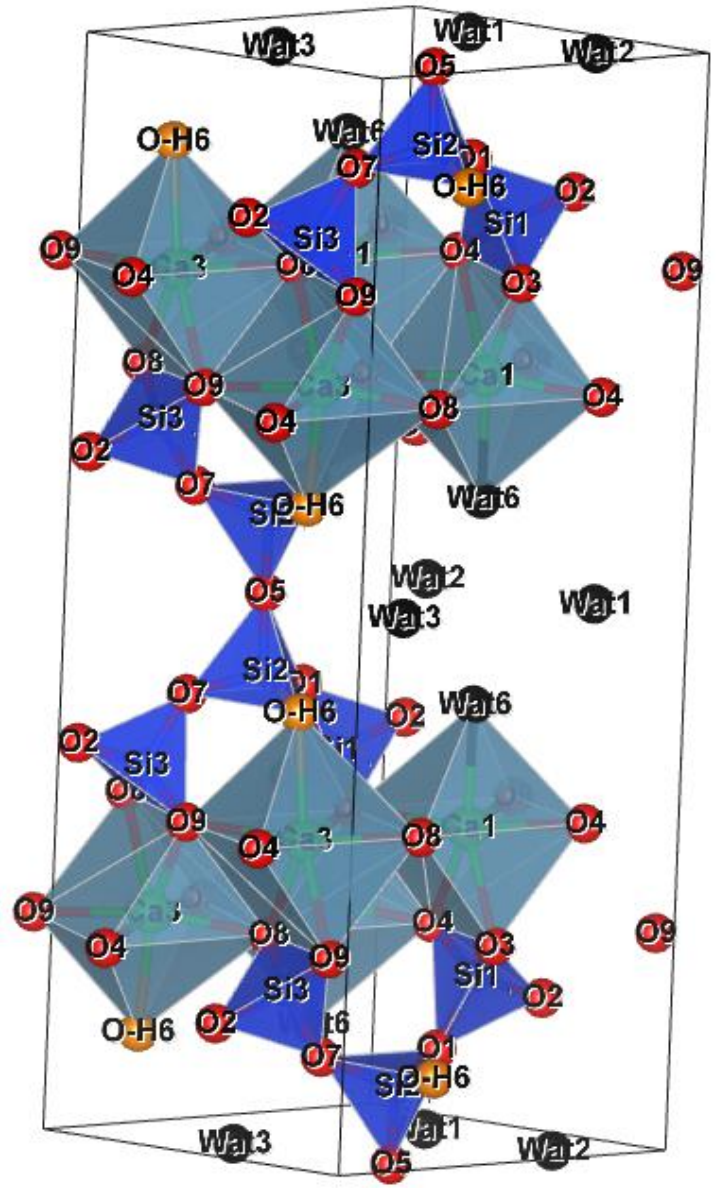
$Ca_5(Si_2O_7)(CO_3)_2$

Tilleyite is a ***soro*carbosilicate** with a very high incorporated CO_2 content (in the form of carbonate groups).

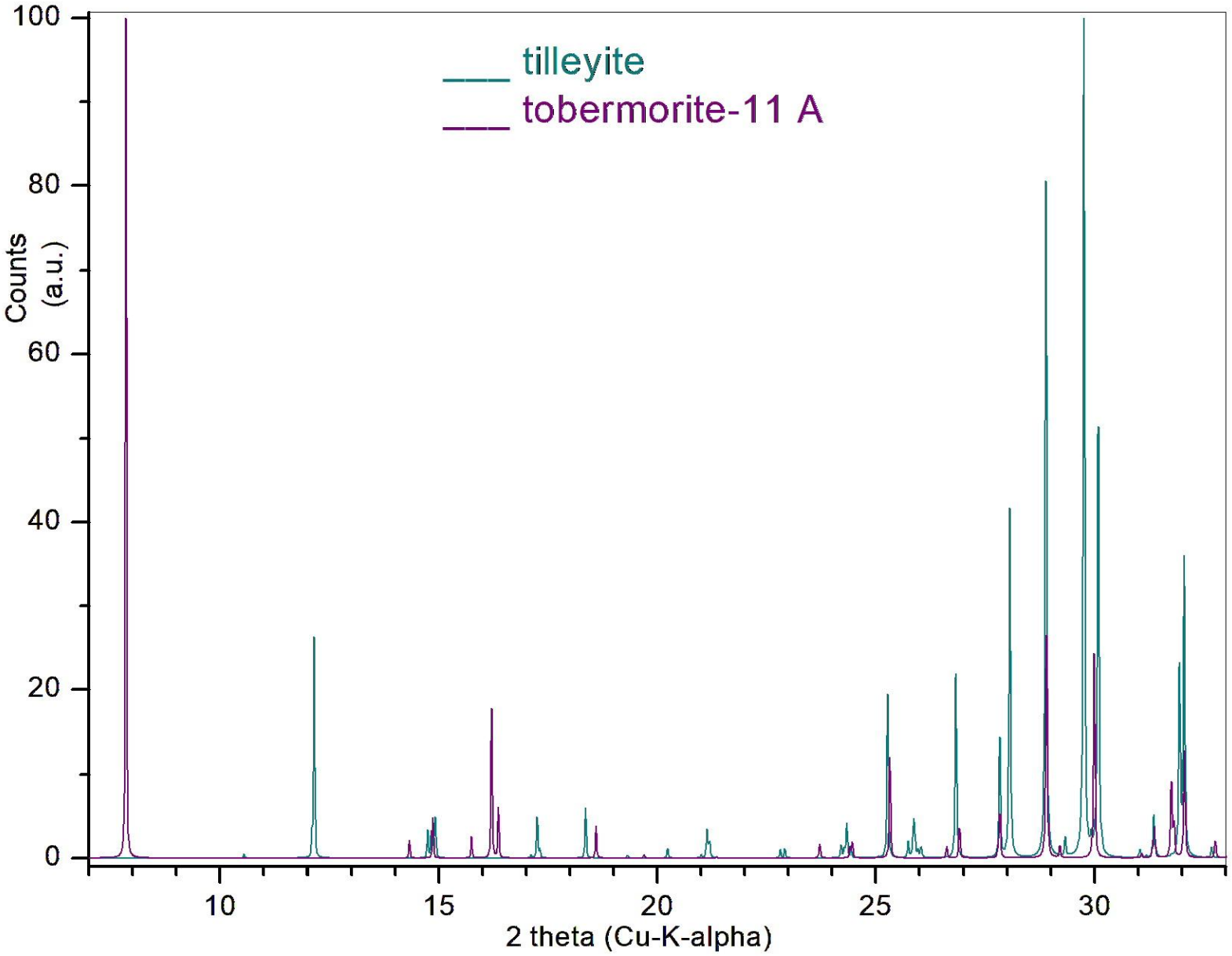
Tobermorite-11 Å, on the other hand, is an **inosilicate**, with unique water molecule insertions into the structure. Waters are positioned at the middle of the unit cell and at the top and the bottom faces of the unit cell.

It will be hard to spot any similarities between the unit cells of tilleyite and tobermorite.

TOBERMORITE – 11 Å



$Ca_{4.5}Si_6O_{16}(OH)\cdot 5H_2O$



Tilleyite			Tobermorite-11 Å		
h k l	2θ	I/I_o	*h k l*	2θ	I/I_o
(2 0 0)	12.151	19	(0 0 2)	7.857	100
(1 2 0)	18.360	5	(0 0 4)	15.751	3
(1 2 -1)	21.138	3	(-1 0 1)	16.210	10
(0 0 2)	24.339	4	(0 -1 2)	16.362	3
(3 2 0)	25.272	18	(0 0 6)	23.721	2
(1 3 0)	26.816	21	(1 0 5)	25.322	8
(3 1 -2)	27.820	13	(-1 2 3)	26.895	2
(1 1 2)	28.052	40	(0 1 6)	27.822	4
(2 3 0)	28.877	80	(-2 2 0)	28.889	10
(4 0 -2)	29.745	100	(0 2 2)	29.984	9
(2 0 2)	30.075	52	(-1 1 7)	31.362	3
(2 1 2)	31.346	5	(-2 0 0)	31.748	7
(5 1 0)	31.934	20	(-1 0 7)	32.049	10
(3 3 0)	32.042	37	(-2 0 2)	32.756	2

The XRD patterns (obtained using Vesta) of tilleyite (of its structure and CIF given in AMCSD 0006060) and tobermorite-11 Å (based on the CIF of AMCSD 0002330) are plotted here. The table provides the most intense X-ray peaks, that will be observed for each phase, over the 2θ range of 7 to 33°.

All six compounds of the previous page are naturally-occurring minerals. Some of them (wollastonite in the Solidia cement [11] and tobermorite in any industrial autoclaved (~180°C at 11.5 atm) aerated concrete) are also produced synthetically at large industrial volumes. Spurrite and tilleyite are carbosilicate compounds (or phases or minerals) that are known [12, 13] to generally form at high temperatures (much higher than 95°C) and high pressures (much higher than the ambient pressure). Spurrite was reported to be encountered as small deposits in a cement kiln preheater co-existing with belite (β-Ca_2SiO_4) [14]. However, L. J. McDonald *et al*. [15] mentioned the possibility of forming trace amounts of scawtite (and tilleyite) at ambient conditions in hydrated Portland cement blended with freshly-precipitated nano-size calcium carbonate upon hydration-curing for 28 days, where the major phases of the cured samples were calcite, $Ca(OH)_2$, ettringite and hemi- and mono-carboaluminates. The McDonald *et al*. study [15] was not able to produce scawtite as the major phase of any sample.

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[11] A. C. Tas, "Mineral additives and production of lightweight composite materials from carbonatable calcium silicate," U.S. Patent No: 11,767,264 B2, September 26, 2023.

[12] D. Santamaria-Perez, J. Ruiz-Fuertes, T. Marqueno, J. Pellicer-Pores, R. Chulia-Jordan, S. MacLeod, and C. Popescu, "Structural behavior of natural silicate-carbonate spurrite mineral, $Ca_5(SiO_4)_2(CO_3)$, under high-pressure, high-temperature conditions," *Inorg. Chem.*, 57, 98-105 (2018).

[13] R. I. Harker, "The synthesis and stability of tilleyite, $Ca_5(Si_2O_7)(CO_3)_2$," *Am. J. Sci.*, 257, 656-667 (1959).

[14] W. Kurdowski and M. Sobon, "Mineral composition of build-up in cement kiln preheater," *J. Therm. Anal. Calorim.*, 55, 1021-1029 (1999).

[15] L. J. McDonald, W. Afzal and F. P. Glasser, "Evidence of scawtite and tilleyite formation at ambient conditions in hydrated Portland cement blended with freshly-precipitated nano-size calcium carbonate to reduce greenhouse gas emissions," *J. Build. Eng.*, 48, 103906 (2022).

The thermal stability of some of the carbosilicates cited in this note was investigated by quite versatile TGA (thermogravimetric analysis) or isothermal heating (at progressively increased temperatures) experiments.

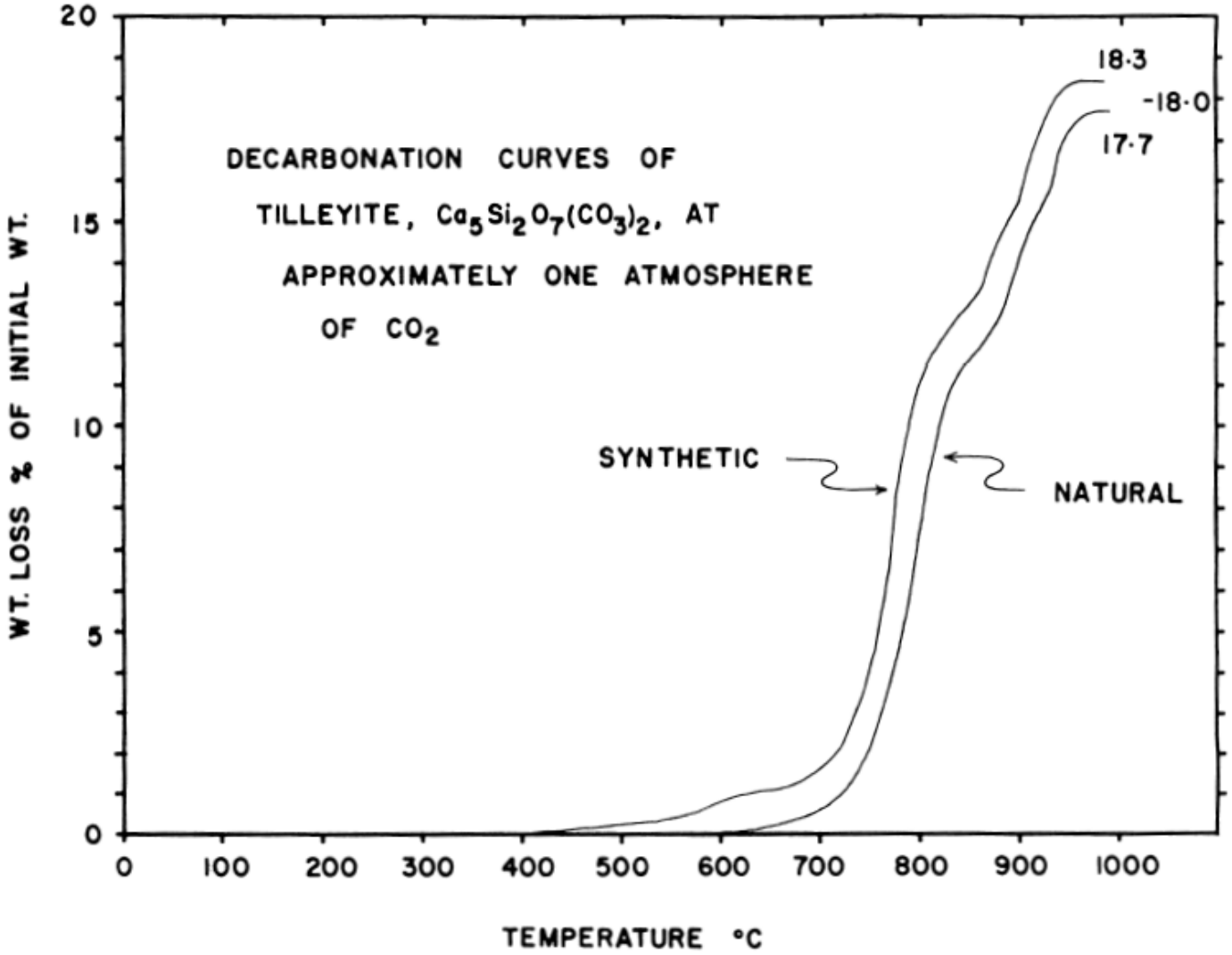
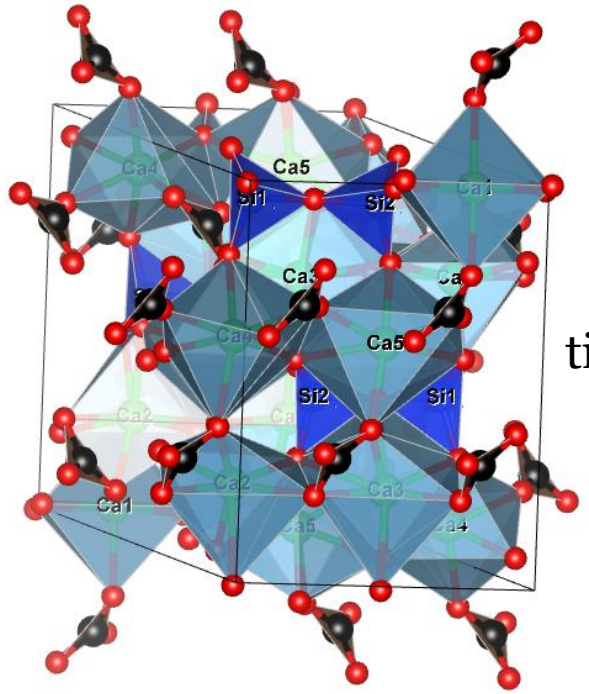
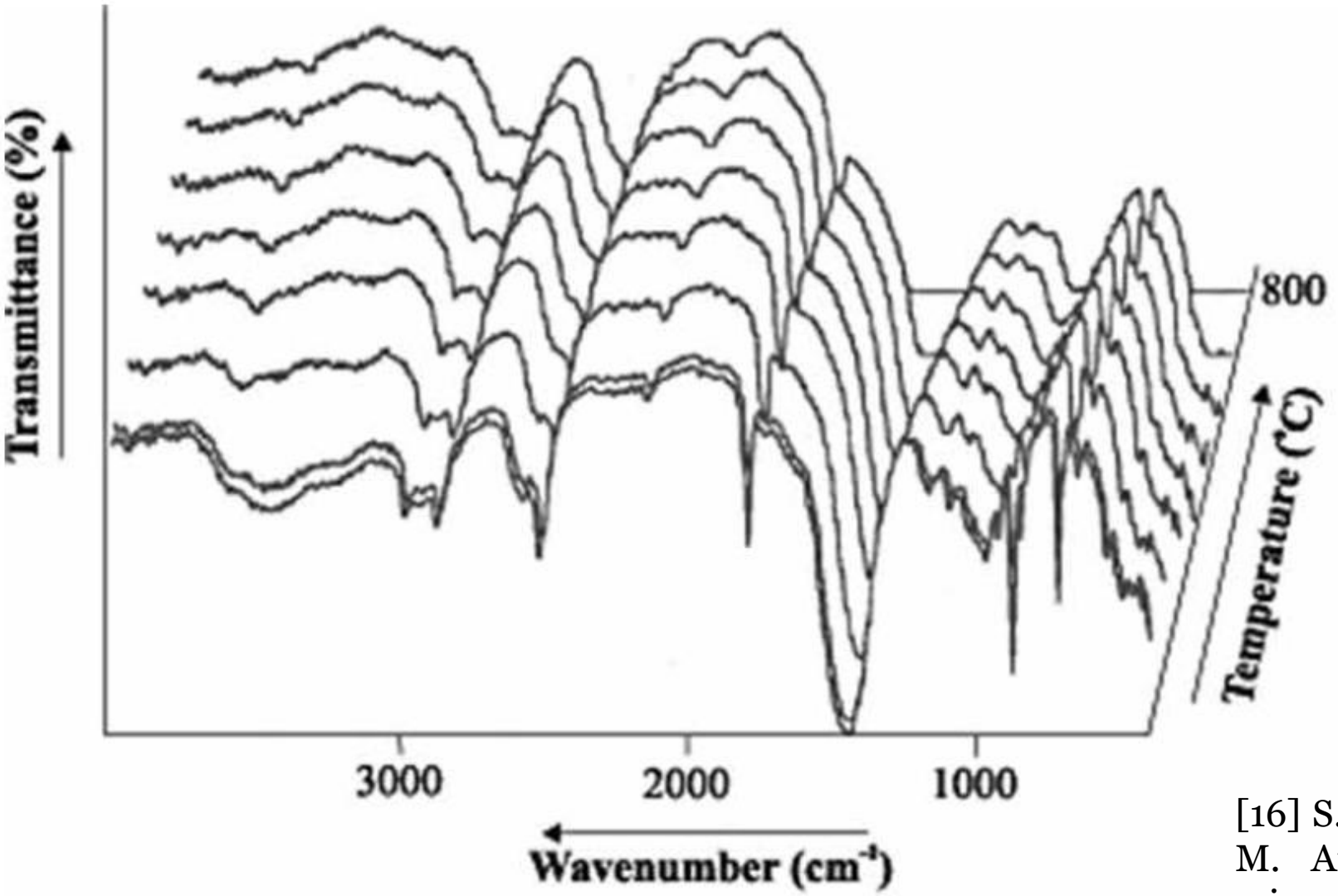


Fig. 1. Thermal dissociation curves of Natural Tilleyite from Carlingford, Ireland, (Material provided by Professor C. E. Tilley), and of Synthetic Tilleyite, (from run 65, see Table 1). The runs were carried out at one atmosphere in a continuously recording thermobalance. Tilleyite ideally contains 18.0% CO_2.

Tilleyite *soro*carbosilicate is known not to decompose easily at temperatures below 600°C. This figure is from reference [13]. The loss of CO_2(g) from tilleyite continued until about 960°C. Pure $CaCO_3$, on the other hand, starts its thermal decomposition at above 550°C and that decomposition into CaO is mostly completed at ~800°C.



tilleyite

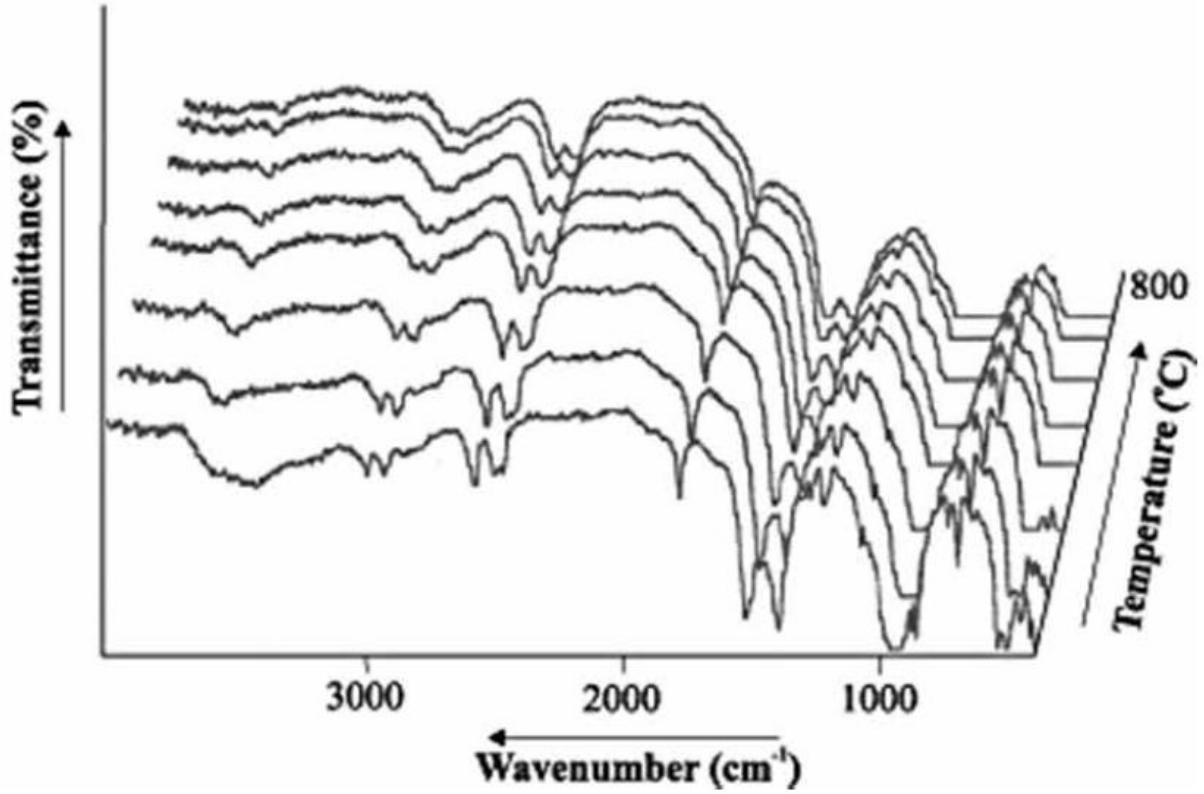


This plot is from Fig, 2 of reference [16]. **Tilleyite** samples were first heated at different temperatures up to 800°C and then their ATR-FTIR (attenuated total reflection-Fourier transform infrared spectroscopy) spectra were collected to show that tilleyite remains stable without any signs of thermal decomposition within that temperature range. This data confirmed what was reported by the study [13] of the previous page.

[16] S. Marincea, D. G. Dumitras, N. Calin, and A. M. Anason, "Spurrite, tilleyite and associated minerals in the exoskarn zone from Cornet Hill (Metaliferri Massif, Apuseni Mountains, Romania)," *Can. Mineral.*, 51, 359-375 (2013).

As of the date of the current note, we were not able to find any data in the open literature on the thermal decomposition of the naturally-occurring carbosilicate mineral galuskinite.

Glasser reported that spurrite did not start its thermal decomposition until 912 ± 5°C [17]. Moreover, the below plot (which is from Fig. 2 of reference [16]) for heated spurrite corroborated Glasser's earlier findings.

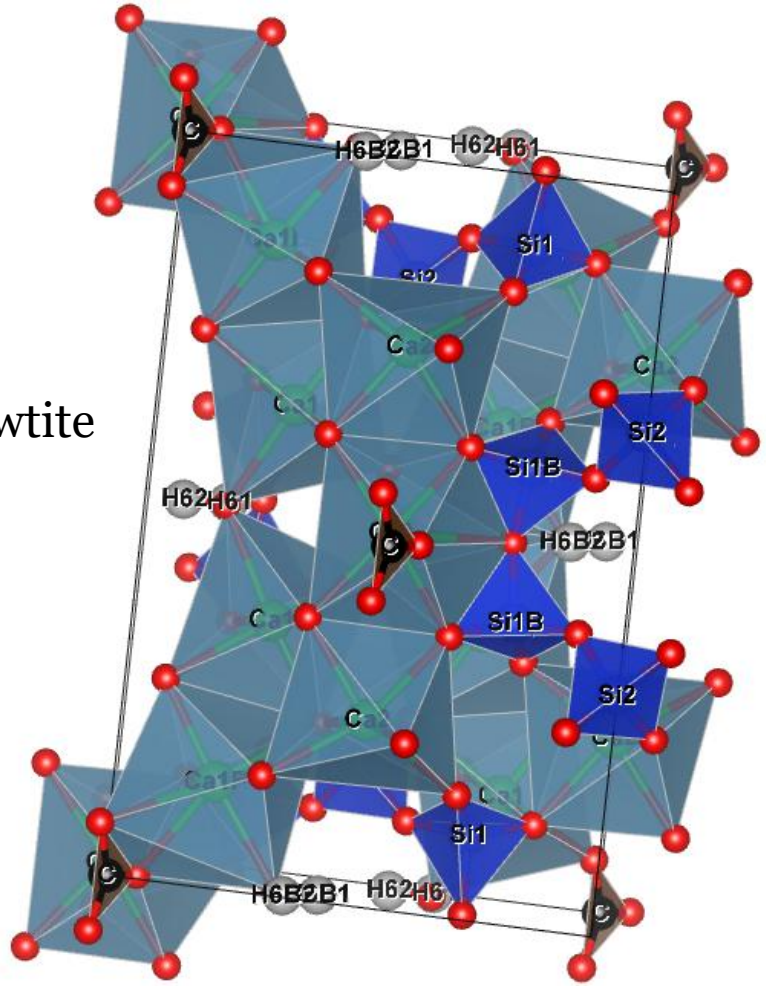


[17] F. P. Glasser, "The formation and thermal stability of spurrite, $Ca_5(SiO_4)_2CO_3$," *Cem. Concr. Res.*, 3, 23-28 (1973).

What is the thermal stability of scawtite?

The article of Maycock and Skalny [18] reported that **scawtite** can be formed during the hydration-curing of C_3S (alite) samples (when the pore solution of such samples contained carbonate ions) and scawtite started its decomposition at around 300°C with a combined evolution of CO_2 gas and mass loss.

scawtite



The below was what we wrote above for scawtite when examining it crystal structure:

"Waters (grey) and CO_3^{2-} groups are located close to the unit cell faces or corners, which typically results in **thermodynamically easier** dehydration-rehydration and decarbonation-carbonation events.

[18] J. N. Maycock and J. Skalny, "Carbonation of hydrated calcium silicates," *Cem Concr. Res.*, 4, 69-76 (1974).

Below, in quotation marks, is an excerpt from the article of Maycock and Skalny [18]. Their study used TG-MS (combined thermogravimetry and mass spectroscopy) as the analytical tool, therefore, they were able to differentiate between the water and CO_2 evolution during heating their samples.

"A likely possibility is that the CO2 evolution peak at 300°C is due to the mineral scawtite, $Ca_7(Si_3O_9)_2CO_3 \cdot 2H_2O$, which is believed to be similar to the phase CSH. This conclusion is based, primarily, on the fact that scawtite has been observed to form in the $CaO\text{-}SiO_2\text{-}H_2O$ system in which slight CO_2 contamination had occurred and that its stability range extends from 140°C to 300°C (18). Inspection of Figs. 1 and 2 also shows that, when the availability of CO_2 or CO_3^{2-} is increased, the CO_2 evolution peak at 300°C increases. If this hypothesis is correct, then unless CO_2 is rigorously excluded from the atmosphere during hydration, scawtite will be present as a hydration product."

The original reference (18) of the above snippet copied from the Maycock and Skalny article is given below as the reference [19] of this note.

- Stevula and Petrovic were able to synthesize relatively high purity scawtite *via* hydrothermal synthesis at 200°C by using CaO, magnesite and quartz mixtures in water as the starting materials [20].

[19] R. I. Harker, "Scawtite and its synthesis," *Mineral. Mag.*, 34, 232-236 (1965).
[20] L. Stevula and J. Petrovic, "Formation of scawtite from mixtures of CaO, magnesite and quartz under hydrothermal conditions," *Cem. Concr. Res.*, 11, 549-557 (1981).

Connection between the carbosilicates and the quantification of CO_2 sequestered by a calcium silicate-based material of any form

- "Form" means a material of any sample size, sample shape, BET surface area, particle size (if the material under consideration is a powder), density and porosity.
- It is quite unfortunate to notice so many times in the literature that researchers run a routine TGA analysis on their materials which underwent some sort of intentional or accidental carbonation and upon observing some unexpected weight loss below 550°C in the data, they are very much inclined to assign that to the presence of "amorphous calcium carbonate" in the samples. The researchers are well-accustomed to read the ASTM standards that strictly specify the $CaCO_3$ decomposition start temperature as 550°C. We think many researchers might not be aware of the fact that amorphous calcium carbonate, if one indeed has it in a sample together with crystalline calcite/aragonite/vaterite, does not necessarily decompose at temperatures below 550°C. The below references [21, 22], which are based on TGA analysis of amorphous calcium carbonate (ACC) samples carried out by competent groups, may be carefully studied by researchers who think that the weight loss below 550°C is due to the presence of amorphous calcium carbonate in the sample.

[21] N. Koga, Y. Nakagoe, and H. Tanaka, "Crystallization of amorphous calcium carbonate," *Thermochim. Acta*, 318, 239-244 (1998).
[22] Y. Kojima, A. Kawanobe, T. Yasue, and Y. Arai, "Synthesis of amorphous calcium carbonate and its crystallization," *J. Ceram. Soc. Japan*, 10, 1145-1152 (1993).

Especially one carbosilicate, *i.e.*, scawtite, we discussed in this note has a thermal decomposition temperature lower than 550°C. Many researchers may be thinking it was ACC in their TGA spectra.

Scawtite

(1) has a low CO_2 content, 5.28 wt% (this means that it can form even when the CO_2 available for its nucleation is not that high),
(2) has a crystal structure in which the CO_3^{2-} groups are located at the corners of its crystallographic unit cell (other carbosilicates do not have this attribute), which makes them easier (i.e., require less energy) to decarbonate,
(3) has quite a low decarbonation temperature (as proved by experimental work),
(4) has the lowest Ca-to-Si molar ratio (1.167) among all the other carbosilicates known, this Ca-to-Si molar ratio is similar to that of C-S-H, therefore, the carbonation of C-S-H is suitable to produce scawtite rather than spurrite, tilleyite or galuskinite.

• The identification of scawtite in a sample requires careful PXRD (powder X-ray diffraction) analysis, this was why we specified the narrow 2θ range of 10 to 33° in p. 11. The field practitioners must run at least 4 h-long "slow" PXRD scans over this narrow range (many researchers scan the 5 to 80° 2θ range in 60 to 90 minutes, then such fast scans generate low resolution PXRD data).
• The identification of any CO_2 (g) generation at any temperature can only be achieved by running either combined TGA-FTIR or combined TGA-MS analyses. The combined TGA-FTIR instruments are always much easier to use. A combined TGA-FTIR-GCMS instrument is the best available tool for a researcher of the field of CO2-sequestering materials.