ponent is the ratio of its distribution by weight between the two phases.

2. The mutual solubility of phenol (m. p., 40.92° ± 0.01) and water has been redetermined between 20° and the consolute temperature; this has been found to be 65.85° ± 0.15° and the consolute concentration is 34% of phenol.

3. The mutual solubility of normal butyl alcohol (b. p., 117.7–117.8°) has been determined between the temperature of the quadruple point, —2.95°, and the consolute temperature, 125.15°; the consolute concentration is 32.5% of *n*-butyl alcohol.

New York, N. Y.

[Contribution from the Chemical Section, Physical Chemical Unit, Pittsburgh Experiment Station, Bureau of Mines, United States Department of Commerce]

THE SOLUBILITY OF CALCIUM SULFATE AT BOILER-WATER TEMPERATURES[1,2]

By R. E. Hall,[3] J. A. Robb[4] and C. E. Coleman[5]

Received January 27, 1926 Published April 5, 1926

The solubility of gypsum has received much attention, and the values are well established. The solubility of those phases of calcium sulfate that may exist at the temperatures of boiler waters has long needed direct determination to substantiate the data of the literature.

The Solid Phase in Equilibrium with the Solution

A number of investigators[6] have described different forms of anhydrous calcium sulfate; van't Hoff and co-workers,[7] however, recognized but two forms, and showed the transition temperatures between them and gypsum to be 66° for gypsum-natural anhydrite, and 89° for gypsum-soluble anhydrite. They assert that gypsum and natural anhydrite are the only forms of calcium sulfate that are stable in the presence of solution, and the

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[1] Published with the approval of the Director, U. S. Bureau of Mines.

[2] This work was done in part in the coöperative investigation of boiler-water conditioning by the Bureau of Mines and the Hagan Corporation of Pittsburgh, Pa., and in part as a fellowship problem of the coöperative arrangement between the Bureau of Mines and the Carnegie Institute of Technology.

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[4] Carnegie Institute Fellow.

[5] Chemist, Hagan Corporation.

[6] Shenstone and Cundall, *J. Chem. Soc.*, **53**, 544 (1888). Lacroix, *Compt. rend.*, **126**, 360, 553 (1898); *Bull. soc. franç. mineral.*, **21**, 39 (1898). Davis, *J. Soc. Chem. Ind.*, **26**, 727 (1907). Rohland, *Z. anorg. allgem. Chem.*, **65**, 105 (1910). De Forcrand, *Bull. soc. chim.*, **35**, 781 (1906). Potilitzin, *Chem. Zentr.*, **1894**, II, 515, 609; **1895**, II, 1066. Cloez, *Bull. soc. chim.*, [3] **29**, 169 (1903). Zunino, *Gazz. chim. ital.*, **30**, I, 333 (1900).

[7] Van't Hoff and co-workers, *Z. physik. Chem.*, **45**, 257 (1903).

conditions under which one or the other form is stable depend on the temperature and nature of the solution with which it is in contact. Thus in water, gypsum is stable up to about 66°; above this temperature, anhydrite is the stable phase. In the presence of other salts, however, the transition temperature may be much lower (for example, 30° in a saturated solution of sodium chloride).

From a study of the volume relations of gypsum and anhydrite it would appear that gypsum should be the stable form under high pressure, for the volume occupied by a given weight of gypsum is less than that occupied by the equivalent weights of water and anhydrite composing it. Farnsworth,[8] however, has pointed out that anhydrite cannot be hydrated under conditions of pressure and temperature analogous to those existing in boiler waters.

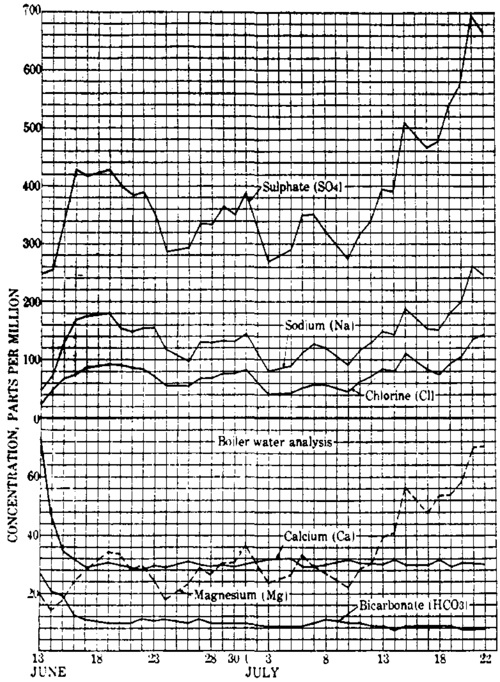

Fig. 1.

For pressures and temperatures characteristic of boiler waters, therefore, it seems definitely enough proved that the solution will be in equilibrium either with soluble or natural anhydrite. In solubility determinations in this temperature range (100° to 225° or higher) we must therefore take careful cognizance of the solid phase in equilibrium with the solution.

In scale formed by boiler waters (at an operating pressure of $6^2/_3$ atmospheres (100 lb.) or more) calcium sulfate, if present, is always in the form of anhydrite.[9] The only indication that has come to our notice of the existence of any other equilibrium was found during the summer of 1922, when we were making a chemical balance test on a boiler.[10] The boiler had been thoroughly cleaned for the test, and the samples of boiler water were carefully filtered and cooled below 100° in a cooling spiral, thus avoiding any changes in concentration. Their analyses are shown in Fig. 1. The only apparent

[8] *Ind. Eng. Chem.*, **17**, 967 (1925).

[9] Hall and Merwin, *Trans. Am. Inst. Chem. Eng.*, **16**, II, 91 (1924).

[10] *Ind. Eng. Chem.*, **17**, 283 (1925).

explanation for the high value of calcium during the first three days is that equilibrium with anhydrite as solid phase was established only slowly and that meantime the solubility corresponded to equilibrium with soluble anhydrite. The scale found in this boiler when it was finally opened was almost pure anhydrite.

Previous Determinations at Elevated Temperatures

Cousté[11] determined the solubility of calcium sulfate in sea water for the temperatures 103° to 133.3°. A solution of the salt of known concentration was introduced into a glass tube, which was then sealed off. The tube and contents were heated in an oil-bath until the solid first began to crystallize. The temperature at this point was noted as that at which the solution was saturated. Cousté found a marked decrease in solubility, but drew the erroneous conclusion that the salt was completely insoluble at 140°. The data are very inexact, and of historical interest only.

Sullivan[12] stated that he proved calcium sulfate to be insoluble in water at 150°, but gave no detail of his experiments.

Tilden and Shenstone[13] made use of a silver-plated, gun-metal tube divided into two parts by a platinum gauze. The capacity of each chamber was 16.4 cc. An excess of precipitated gypsum was introduced with water into one end of the tube, which was then closed and maintained at the desired temperature. After a suitable period of heating, the tube was inverted, and the solution filtered from excess solid by passage through the gauze. After cooling, the tube was opened and the solution analyzed. Tilden and Shenstone's results expressed as grams of calcium sulfate per million grams of solution are in Table I.

TABLE I

RESULTS OF TILDEN AND SHENSTONE'S DETERMINATIONS

Temp., °C	140	165	180	240	250
$CaSO_4$, p.p.m.	779	560	270	180	180

Doubtless the solid phase at temperatures below 200° was soluble anhydrite, as precipitated gypsum was used in the beginning. On the whole, the accuracy of this work does not compare with that noted below.

Boyer-Guillon[14] made use of a beautifully constructed bronze autoclave of 20 liters' capacity. In this was placed a solution of calcium sulfate in distilled water, with an excess of solid phase present, presumably gypsum, though not specified by the author. The temperature was raised to the desired point and maintained constant as nearly as possible until equilib-

[11] Cousté, *Ann. des Mines*, **5**, 140 (1854).

[12] Sullivan, *British Assoc. Repts.*, **1859**, 291.

[13] Tilden and Shenstone, *Phil. Trans. Roy. Soc.*, **175**, Pt. 1, 23 (1884).

[14] Boyer-Guillon, *Ann. Conserv. Arts Metiers*, [3] **2**, 189 (1900).

rium was attained. Samples of 100 cc. each were withdrawn through an internal asbestos filter, and finally through an external cooling coil. Equilibrium was attained slowly (more than eight hours at 100°; one hour at 200°); its attainment was insured by taking samples every hour or half hour, plotting concentration of calcium sulfate in relation to time, and continuing the experiment until the curve was parallel to the time axis.

The results in grams of calcium sulfate per million grams of solution are as in Table II.

Table II

Results of Boyer-Guillon's Determinations

Temp., °C........	100	119.7	134.0	151.2	169.6	200.4
$CaSO_4$, p.p.m.....	1649	1008	699	486	310	155

The main criticism of Boyer-Guillon's work is his omission of blowing off steam at intervals. As the crystals of calcium sulfate settle readily, this omission means an absence of solid phase in the vicinity of the filter, and might permit some supersaturation.

In 1910, Melcher[15] published his work on indirect solubility determinations of slightly soluble salts by means of conductance measurements in the platinum-lined steel bomb described by Noyes and Coolidge.[16] He was the first to note the different solubility values obtainable by starting with gypsum or anhydrite as solid phase in contact with the solution. His anhydrite was prepared by bringing precipitated gypsum to constant weight at 200° (12 hours of heating required). When he started with gypsum as the solid phase, he obtained one set of solubility values, so long as the solution was not heated above 110°. When the temperature was raised to 135° and then lowered to 100°, a lower value was obtained than with gypsum at this temperature. This value, which represents the solubility of soluble anhydrite, is in good agreement with van't Hoff's figures. When this solution was heated to 218°, however, and then cooled to 156° or 100°, the solubility of the calcium sulfate was no longer that of gypsum or soluble anhydrite, but corresponded to that of anhydrite.

The results for gypsum were obtained by raising the temperature of the bomb containing gypsum and water from room temperature to that desired, and then maintaining it at this point until equilibrium was attained. The results for soluble anhydrite were obtained by heating to 135–161°, and then maintaining the desired (lower) temperature until equilibrium was certain. The results for anhydrite were obtained as for gypsum, anhydrite being the solid phase introduced into the bomb with the solution. Melcher's results[17] are in Table III.

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[15] Melcher, This Journal, **32**, 50 (1910).

[16] Noyes and Coolidge, *Carnegie Inst. Pub.*, **63**, 59 (1907).

[17] The calculated values quoted in Landolt Börnstein "Tabellen" [5th ed., p. 645] have failed to recognize the temperature at which the concentration was measured.

TABLE III

RESULTS OF MELCHER'S DETERMINATIONS

Temp., °C	18	50	100	156	218
Gypsum ($CaSO_4$), p.p.m.	2008	2064	1653	...	...
Soluble anhydrite, p.p.m.	..	..	1618	478	...
Anhydrite, p.p.m.	..	..	653	202	57

As Melcher's results for soluble anhydrite check Boyer-Guillon's results very closely, it seems certain that the latter was determining the solubility of this form of calcium sulfate.

The Solubility of Anhydrite as Determined by Direct Methods

We shall now present the results of the present investigation.

Apparatus and Procedure

Autoclave and Filter.—For our determinations, we made use of an iron autoclave (Fig. 2) of about 7 liters' capacity, which was provided with an internal filter *f*, and a thermometer well *t*. The filter consisted of 10 layers of finely woven cloth, supported by perforated metal plates which were firmly fastened in a steel supporting frame (Fig. 3). On leaving the autoclave the filtrate passed through a copper cooling coil in which it came practically to room temperature. Thus change in concentration by evaporation was avoided. The temperature was controlled by a carefully standardized copper-constantan thermocouple *p*, inserted at *t*. Its e.m.f. was measured on a Type K[18] potentiometer. Steam was blown off through *s* as desired.

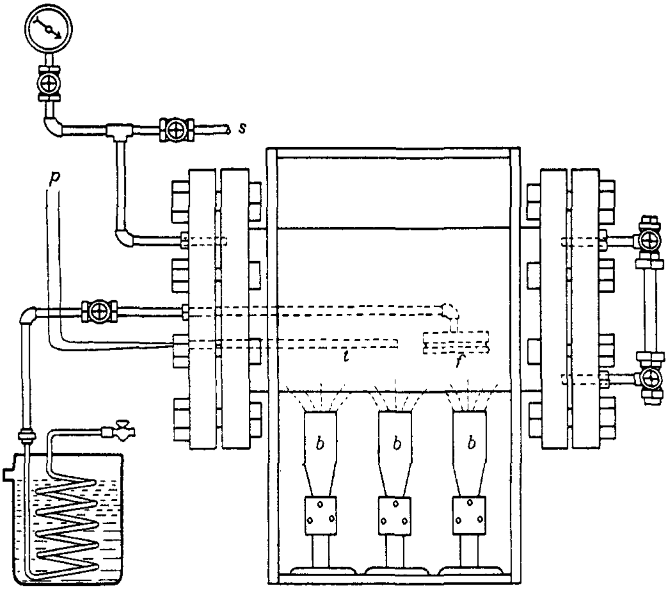

Fig. 2.—Autoclave.

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[18] Leeds and Northrup Co., Catalog 70, 1922.

Temperature Control.—The temperature of the autoclave could be held satisfactorily constant at the higher temperatures, but at the lower the maintenance of a constant temperature was more difficult. As a small fluctuation under the latter condition gave rise to quite a large difference in solubility, the results at the lower temperatures show greater divergence from a smooth curve than those at the higher.

Solid Phase.—For determinations of the solubility of soluble anhydrite, $CaSO_4.2H_2O$ was the solid phase introduced; for determinations of anhydrite, anhydrite was the solid phase; for while anhydrite is the stable solid phase above 66°, according to van't Hoff, that obtained when gypsum is the solid phase introduced is soluble anhydrite for temperatures approaching 200°. Above 200°, the transformation to anhydrite seems to occur readily enough (see Melcher, and Expts. 29 and 32, Table V). Several microscopical examinations of the solid phase remaining in the autoclave after a determination in which anhydrite was the solid phase introduced, were made by De Chicchis,[19] who found them to be anhydrite in all cases.

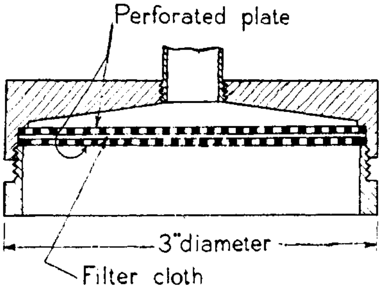

Fig. 3.—Internal filter.

The calcium sulfate used was the regular C. P. $CaSO_4.2H_2O$ of the analytical laboratory. The anhydrite was prepared by igniting this at a dull red heat in a muffle furnace for from three to six hours. A solution of the ignited material in distilled water free from carbon dioxide was neutral to phenolphthalein, thus indicating that no decomposition into calcium oxide and sulfur trioxide occurred.

Procedure.—The detailed procedure in each determination is given in the summary in Table V. Experience gained in the work showed that the most consistent results could be obtained as follows. Six liters of distilled water was introduced into the autoclave and heated to 100°; 10 to 20 g. of anhydrite was added, the autoclave was closed, the temperature raised to that desired, and maintained until the samples for analysis were withdrawn. Steam was blown off every hour during the experiment so as to agitate the solution, but not within at least an hour before sampling. Immediately after sampling, the autoclave was completely drained to prevent any reversion of anhydrite to gypsum on standing in contact with water at room temperature.

Samples.—When samples were taken, the first 200 cc. of filtrate was discarded. Only two 1-liter samples could be taken per experiment.

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[19] R. De Chicchis, Fellow at the Carnegie Institute of Technology and the Bureau of Mines, investigating mineral dusts, 1924–25.

However, different runs were made at the same temperature but for various periods, so that agreement in concentration indicated that equilibrium had been attained.

The samples were measured in 1-liter volumetric flasks at 18°. The densities of the various samples were not determined but were taken from a straight-line curve obtained by plotting density against grams of calcium sulfate per 100 cc., 1.00059 being taken as the density[20] of a solution containing 0.2116 g. of calcium sulfate per 100 cc. and 0.99862 as the density of pure water at 18°. Consideration of this curve showed that the error introduced by assuming the density equal to 1.0 was well within the limit of error of the determination; consequently, in calculating from grams per liter to parts per million of solution, we made this assumption.

It might be thought that the boiler surfaces with which the solution came in contact during heating might be detrimental to its purity. Tests made by Coleman, however, showed that the grams per liter of calcium sulfate, calculated from determinations of both calcium and sulfate, checked very closely the total solids obtained by evaporation, as in Table IV.

TABLE IV

COMPARISON OF RESULTS BY ANALYSIS AND EVAPORATION

$CaSO_4$, by analysis	0.0264	0.0662	0.1014	0.0600	0.1129
$CaSO_4$, by evaporation	.0296	.0680	.1008	.0625	.1143

On concentration, the samples became slightly yellow in color, which became more intense when the solution was made alkaline; but it remained soluble and was readily washed from the precipitated calcium oxalate.

The Analysis.—The 1-liter samples, acidified with nitric acid, were concentrated in Pyrex glass to 100 cc. Ammonia was added until the solution was alkaline. Any iron that precipitated was filtered off and if appreciable in amount a re-precipitation was made. The combined filtrates and washings were concentrated to 200 cc. To the boiling solution an excess of saturated ammonium oxalate solution (25 to 50 cc.) was added. After boiling gently for five minutes the solution was removed from the hot-plate, made definitely alkaline with ammonia, and allowed to stand overnight. The precipitate was then filtered off, and washed with dil. ammonia water until free from oxalate. The beaker was thoroughly washed with ammonia water, but was not scrubbed with a policeman. The filter paper was removed from the funnel, opened and laid against the inside of the beaker (free from soluble oxalate) in which the precipitation was made. The precipitate was washed from the paper into the beaker, and 100 cc. of boiling distilled water plus 5 cc. of 1:1 sulfuric acid was added. The solution was then titrated with 0.05 *N* potassium permanganate solution. At the first permanent pink, the paper was added, and

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[20] Hulett and Allen, THIS JOURNAL, **24**, 667 (1902).

titration continued until a permanent pink was obtained. The permanganate solution was standardized at intervals against sodium oxalate certified by the United States Bureau of Standards.

Data obtained by the writers are given in Table V.

TABLE V

DATA ON SOLUBILITY OF ANHYDRITE

Expt.	Conditions	Total time at temp., hours	$CaSO_4$, p.p.m.
	At 100°		
1	Anhydrite made by heating gypsum for $^1/_2$ hr. at 300–400°, introduced into boiler with distilled water and allowed to stand for 2 days before the determination. Samples, 500 cc.	5 8	(1150) (1180)
2	Anhydrite made by heating gypsum in muffle furnace at 200–600° for 5 hrs., added to boiling distilled water in the boiler. Samples, 90–125 cc. The anhydrite was examined microscopically by W. M. Myers,[a] and found to be completely dehydrated.	$3^3/_4$ 5	786 835
	At 116°		
3	Anhydrite, made as in Expt. 2, introduced into boiling water, temp. held at 162° for $^1/_4$ hr., then lowered to 116° and held constant. Occasional blow-off. Sample, 110 cc.	4	711
	At 122°		
4	Anhydrite (12 g.), made by heating gypsum to a dull red heat for 3 hrs., was added to boiling water (6 liters) maintained at 100 lb. gage pressure for 1 hr., then lowered to 122°. Blow-off every hour. Samples, 1 liter.	$8^1/_2$ $12^1/_2$	450 470
5	Anhydrite (15 g.) as in Expt. 4 was added to boiling water (6 liters); boiling was continued for 1 hr. Temp. was then raised to 122°. Blow-off every hour. Samples, 1 liter.	9 (two samples)	670 680
6	Anhydrite (15 g.) as in Expt. 4 added to boiling water (6 liters); temp. raised to 122° and maintained there. No blow-off. Samples, 1 liter.	12 14 16	580 590 600
	At 142°		
7	Anhydrite made as in Expt. 1 was introduced into the boiler with distilled water and left standing at room temperature for two days previous to the determination. Then the boiler was held at 100 lb. gage pressure for 4 hrs., then dropped to 146°, where it was held constant. No blow-off. Samples, 500 cc.	$1^1/_2$ 4	(350) (330)
8	Anhydrite (10 g.) made as in Expt. 4 was introduced into the boiler with distilled water (5 liters), and immediately heated to 145°, where it was held constant. No blow-off. Samples, 1 liter.	$6^1/_2$ $9^1/_2$	350 360

[a] Assistant mineral technologist, U. S. Bureau of Mines.

TABLE V (*Continued*)

Expt.	Conditions	Total time at temp., hours	$CaSO_4$, p.p.m.
9	As in 8, except temperature maintained at 142°. Blow-off every hour. Samples, 1 liter.	6 (two samples)	300 270
10	As in 9, except anhydrite was added to boiling water. Blow-off every hour. Samples, 1 liter.	9	340
		11	340
		13	360
11	Anhydrite (15 g.), made as in Expt. 4, was introduced into boiling distilled water (6 liters). It was boiled for 1 hr. at 100°, then raised to 142° and maintained constant. No blow-off. Samples, 1 liter.	9 (two samples)	410 410
	At 156°		
12	Anhydrite, made as in Expt. 2, was introduced into boiling distilled water; the temp. was raised to 140°. At the end of 2 hrs. a sample of salt was obtained through the gage glass, the water was drained off, alcohol was added twice and decanted, and finally the salt was dried with ether. It was examined with the petrographic microscope by W. M. Myers, who found the lowest index to be 1.550, and the highest to be 1.600, thus indicating total absence of gypsum. After the sample was taken, the temp. of the boiler was raised to 156°, where it was maintained constant. Occasional blow-off. Samples, 100 cc.	5	295
13	As in Expt. 10, except the temp. was maintained at 156°. Blow-off every hour. Samples, 1 liter.	6 (two samples)	230 230
14	As in Expt. 13.	10	260
		12	260
		14	250
	At 166°		
15	Anhydrite, made as in Expt. 1, was introduced into the boiler with distilled water and allowed to stand for 2 days before the determination. The temp. was raised to 172°, and maintained there. No blow-off. Samples, 500 cc.	1½	(240)
		4	(230)
16	Anhydrite (5 g.), made as in Expt. 4, was introduced into the boiler with distilled water (5 liters). The temperature was raised to 168° and maintained constant. No blow-off. Samples, 1 liter.	4	220
		5½	210
17	As in Expt. 10, except the temp. was maintained at 166°. Blow-off every hour. Samples, 1 liter.	6 (two samples)	180 180
18	As in Expt. 17.	9	190
		13	180
	At 175°		
19	As in Expt. 10, except the temp. was maintained at 175°. Blow-off every hour. Samples, 1 liter.	6 (two samples)	150 150
20	As in Expt. 19.	13 (two samples)	150 150

TABLE V (*Concluded*)

Expt.	Conditions	Total time at temp., hours	$CaSO_4$, p.p.m.
	At 182°		
21	As in Expt. 15, except the temp. was maintained at 190°. No blow-off. Samples, 500 cc.	$3\frac{1}{2}$ $4\frac{1}{2}$	(170) (170)
22	As in Expt. 10, except the temp. was maintained at 182°. Blow-off every hour. Samples, 1 liter.	6 (two samples)	120 120
23	As in Expt. 22.	$8\frac{1}{2}$ (two samples)	120 120
	At 195°		
24	As in Expt. 15, except the temp. was maintained at 190° for $4\frac{1}{2}$ hrs., then raised to 204°, and maintained constant. No blow-off. Samples, 500 cc.	$2\frac{1}{2}$ 4	(140) (150)
25	As in Expt. 10, except the temp. was maintained at 195°. Blow-off every hour. Samples, 1 liter.	6 (two samples)	84 83
26	As in Expt. 25.	8 (two samples)	93 94
	DATA ON SOLUBILITY OF SOLUBLE ANHYDRITE, EXCEPT EXPTS. 29 AND 32 WHICH REPRESENT VALUES FOR ANHYDRITE		
	At 115°		
27	A strong solution of calcium sulfate, containing $CaSO_4.2H_2O$ as the solid phase, was introduced into the boiler. Temperature was brought to 115° and maintained constant. Occasional blow-off. Samples, 100 cc.	$4\frac{1}{2}$	1124
	At 122°		
28	As in Expt. 27, except the temperature was maintained constant at 122°.	$4\frac{2}{3}$	1014
	At 123°		
29	As in Expt. 27, except the temp. was maintained at 207° for $1\frac{1}{4}$ hrs., then lowered to 123° and held constant. The results check Melcher's observations, in that change to equilibrium with natural anhydrite has occurred. The solubility checks closely that obtained by Robb for this temp. when he started with anhydrite as solid phase 4, 5, 6.	2	590
	At 142°		
30	As in Expt. 27, except the temp. was maintained at 186° for 1 hr., lowered to 142° and held constant.	$3\frac{3}{4}$	650
	At 186°		
31	As in Expt. 27, except the temp. was maintained at 186°. Nos. 30 and 31 represent one experiment with the boiler.	1	264
	At 207°		
32	As in Expt. 27, except the temp. was maintained at 207°. Nos. 29 and 32 represent one experiment with the boiler. From the solubility values, it is evident that equilibrium is established with anhydrite.	$1\frac{1}{4}$	65

TABLE VI

FINAL RESULTS OF HALL, ROBB AND COLEMAN'S DETERMINATIONS

Temp., °C.	No. of expt. used in av.	$CaSO_4$, p.p.m.	Temp., °C.	No. of expt. used in av.	$CaSO_4$, p.p.m.
	ANHYDRITE			SOLUBLE ANHYDRITE	
100	2	810	115	27	1124
116	3	711	122	28	1014
122	4, 5, 6; 29 at 123°	565	142	30	650
142	8, 9, 10, 11	349	186	31	264
156	12, 13, 14	259			
166	16, 17, 18	196			
175	19, 20	150			
182	22, 23	120			
195	25, 26	89			
207	32	65			

It is apparent that the data in brackets in Col. 4, in which the solid phase was gypsum, heated for one-half hour to a temperature of 300° to 400°, should be discarded. By averaging the other determinations, we obtain the final data given in Table VI.

Our data for anhydrite differ somewhat from Melcher's at the lower tem-

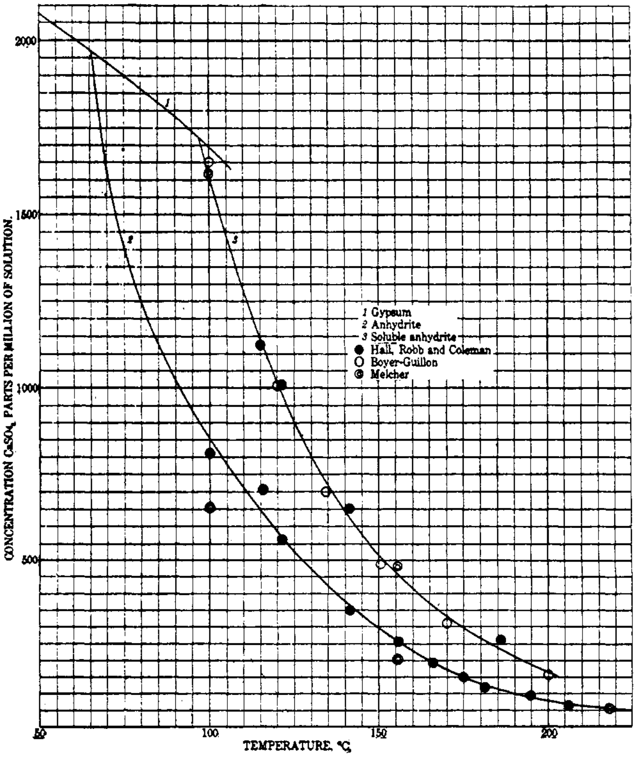

Fig. 4.—Solubility of gypsum, anhydrite and soluble anhydrite.

peratures, but agree well with his at the higher temperatures. The values for anhydrite at rounded temperatures presented in Table VII were obtained by passing a smooth curve through our points and the 218° point of Melcher (Fig. 4). The values for soluble anhydrite were obtained from a smooth curve the position of which was determined by the data of Boyer-Guillon, Melcher and the writers.

TABLE VII

SOLUBILITY OF ANHYDRITE AND SOLUBLE ANHYDRITE AT ROUNDED TEMPERATURES (PARTS PER MILLION)

Temp., °C.	100	120	140	160	180	200	220
Anhydrite	850	587	372	226	130	75	56
Sol. anhydrite	1630	1015	640	410	265	165	...

Summary

Direct determinations have been made of the solubility of anhydrite and soluble anhydrite at temperatures characteristic of boiler waters. The values for soluble anhydrite agree with those of Boyer-Guillon and Melcher; those for anhydrite agree well with Melcher's values at higher temperatures, but not so well with those at lower temperatures.

PITTSBURGH, PENNSYLVANIA

[CONTRIBUTION FROM THE KENT CHEMICAL LABORATORY OF THE UNIVERSITY OF CHICAGO]

THE ISOTOPIC COMPOSITION AND ATOMIC WEIGHT OF CHLORINE FROM METEORITES AND FROM MINERALS OF NON-MARINE ORIGIN

(Papers on Atomic Stability)

BY WILLIAM D. HARKINS AND S. B. STONE

RECEIVED FEBRUARY 1, 1926 PUBLISHED APRIL 5, 1926

1. Introduction. Constancy of the Isotopic Composition of the Element

According to the Whole Number rule of Harkins and Wilson[1] the atomic weights of individual atomic species are whole numbers, with a precision of about 0.1%. On this basis the present atomic weight of ordinary chlorine, 35.457, indicates a composition of 77.15% of chlorine of isotopic number 1, and 22.85% of isotopic number 3.

The investigation described here was undertaken in order to determine to what extent the constancy of composition previously observed in chlorine of marine origin, which might be explained as due to mixing, is exhibited in the chlorine of the meteorites, which has probably never been thoroughly mixed with the terrestrial element.

As a secondary feature of the work the atomic weight of the element

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[1] Harkins and Wilson, THIS JOURNAL, **37**, 1367 (1915).