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EXPANSION AND SHRINKAGE OF OVERSULPHATED PORTLAND CEMENTS

D. W. Hobbs
Materials Research Department, Cement and Concrete Association
Wexham Springs, Slough, Bucks, U.K.

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ABSTRACT

Expansion and shrinkage tests on mortars made from a range of cements with SO_3 contents in excess of the limits specified in BS 12 are described. The mortar specimens were cured under water from 24 hours to 7 days and then dried at 65% RH and 20°C. It is shown that there is a small range of SO_3 contents within which sharp increases in 7 day expansion occur ranging from 80 to 170 micro-strain/SO_3 % and that within this small range of SO_3 contents the subsequent drying shrinkage is not adversely affected.

Quell- und Schwindversuche mit Mörtelproben aus unterschiedlichen Zementarten, und mit einen SO_3-Gehalt, der über den in BS12 angegebenen Grenzwerten lag, werden beschrieben. Die Mörtelproben wurden zwischen 24 Stunden und 7 Tagen unter Wasser gelagert und anschliessend bei 20°C und 65% r.F. getrocknet. Innerhalb eines engen Bereiches des SO_3-Gehaltes wird ein starkes Ansteigen der 7-Tage-Quell-Verformung von 80-170 µD/% SO_3 beobachtet. Im selben engen Bereich des SO_3-Gehaltes wird das nachfolgende Schwinden nich nachteilig beeinflusst.

Introduction

Concrete shrinks initially due to thermal contraction and subsequently due to drying. Because concrete can only support low induced tensile strains, tensile cracking may be induced if this shrinkage is restrained. However in recent tests the author has found that restraint of shrinkage by reinforcement levels as high as 11 per cent does not cause drying shrinkage cracks to initiate in concrete (1). Consequently some of the cracking which is attributed to drying shrinkage may have originated from restraint to thermal contraction.

The shrinkage cracking which occurs in concrete can be reduced or eliminated by using prestressing or a shrinkage-compensated-cement. The shrinkage compensated cements currently in use are (2,3):-

Type K - Portland cement, anhydrous calcium alumino-sulphate, calcium sulphate, free lime.

Type M - Portland cement, calcium-aluminate cement, calcium sulphate.

Type S - High calculated C_3A Portland cement, calcium sulphate.

Type'L'- Portland cement, lime.

In Types K, M and S the expansive force is produced by the formation of ettringite which results from the reaction of C_3A and calcium sulphate, whilst in Type 'L' the expansive force is produced by a hydrating lime system. With these cements a major part of the expansive compound forms after the concrete has acquired some strength and consequently, to prevent undesirable expansions occurring subsequently during the life of the structure, the concrete must be adequately cured so that the expansive reactions are completed by an age of about 7 days.

In the present paper results of expansion and shrinkage tests on cements with SO_3 contents in excess of the limits specified in BS 12 are described. The cements cannot be regarded as Type S expansives because their C_3A contents are too low, nevertheless it is suggested that the small induced expansions may balance, in part, the early age contraction, both thermal and drying, and that the general conclusions may be applicable to Type S expansive cements.

Very few published results are available for Type S expansives. In 1966 Gustaferro, Greening and Klieger (4) deduced from published work on sulphate resistance (5) that a Portland cement with a high C_3A content could be modified to become an expansive cement by raising the SO_3 content sufficiently. Unfortunately in their own tests the SO_3 contents were set too high and as a result the expansions occurred over too long a period (3 weeks). In a later paper Klieger and Greening (6) reported the results obtained on a cement with a C_3A content of 17.1 per cent and an SO_3 content of 4.48 per cent. With this cement the expansive reactions were complete at 7 days and the observed expansion of concrete prisms at 7 days was approximately 1400 micro-strain.

The Cements

Ten cements were tested. Three cements were obtained by grinding clinker in a laboratory ball mill, each clinker being ground to three finenesses, and the remaining seven cements were cements as delivered by the manufacturer. The potential compound compositions of the clinkers and cements obtained from a chemical analysis together with their approximate finenesses are given in Table 1. A complete chemical analysis and a quantitative X-ray diffraction analysis were also carried out on each cement. The finenesses of the cements ranged from 230 to 725 m^2/kg and the C_3A contents varied from 0 to 12 per cent.

To produce cements with a range of SO_3 contents 'hemihydrate'* with a fineness of 624 m^2/kg was added to each of the cements. The hemihydrate was dispersed uniformly in the cement by shaking the mixture vigorously in a large container.

TABLE 1

Potential Compound Composition of Clinkers and Cements Obtained by Chemical Analysis and the Approximate Fineness of the Cements+

'Cement'	C_3S (%)	C_2S (%)	C_3A (%)	'C_4AF' (%)	$CaSO_4$ (%)	Approximate Cement Fineness (m^2/kg)
A1	64	14	0	18	0	315
A2	64	14	0	18	0	400
A3	64	14	0	18	0	515
B1	59	17	11	8	0	250
B2	59	17	11	8	0	350
B3	59	17	11	8	0	430
C1	73	9	7	7	0	230
C2	73	9	7	7	0	355
C3	73	9	7	7	0	440
D	49	18	7	10.6	6.6	725
E	59	20	12	1	3.8	355
F	56	16	9.4	6.4	4.2	375
G	48	23	9	5.5	5.1	440
H	41	34	8.5	5.8	4.1	345
I	60	13	10.2	5.6	4.0	320
J	52	18	6.2	11.8	4.1	365

Experimental Procedure

Preparation of mortar specimens

The mix used throughout this work had a free water/cement ratio of 0.47 and an aggregate/cement ratio of 3.89. The aggregate was a Thames Valley flint sand and the mortar specimens, which were 25 x 25 x 250 mm, were prepared using the normal C and C A procedure (7).

Expansion measurements

After demoulding at 24 hours 5 or 6 specimens from each batch were placed in polythene jackets filled with water at 20°C (20 to 25 g of water was added to each jacket). The specimens were then set up in measuring rigs (see Figure 1) and the axial expansions measured with inductive displacement transducers that have a linear range of 250 µm. The demodulated transducer

* SO_3 content of pure hemihydrate is 55.16 per cent. The SO_3 content of the 'hemihydrate' used in this investigation was 53.7 per cent.

+ The fineness increased slightly with increasing SO_3 content.

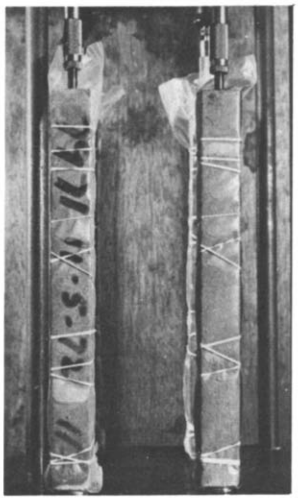


FIG. 1

Specimens Set Up for Expansion Measurements

FIG. 2

Measuring Stand for Shrinkage Specimens

outputs were scanned sequentially for a period of 6 days and displayed on a chart recorder. The chart output was sensitive to a movement of 2 micro-strain. The measuring and recording apparatus has been described in detail by Mears (8,9).

Shrinkage measurements

At an age of 7 days the specimens were set up in measuring rigs (see Figure 2) and the axial shrinkage measured with inductive displacement transducers whilst the specimens were drying for a period of 28 or 105 days in an atmosphere at 65 ± 2% RH and 20 ± 1°C. The transducers had a linear range of 500 µm and the ac outputs from a hundred of these transducers were displayed on a Farnell Digital voltmeter via a 100-channel switching unit. Each transducer was set to operate on the linear part of its travel by its own potentiometer and its sensitivity was adjusted by a second potentiometer so that a change in measured output of 1 millivolt corresponded with a change in specimen length of 2.5 µm. The measuring apparatus was sensitive to a shrinkage movement of 1 micro-strain.

Results

Expansion

The expansion as the mortar specimens made from cements B and D were cured under water from 1 to 7 days is shown plotted in Figures 3a to 3d. In Figures 4a to 4f the total expansion that occurred between 1 and 7 days is shown plotted against SO_3 content. Each point plotted is the mean of 4 to 6 observations; an indication of the standard error of the mean is given in Figures 4b to 4f.

It can be seen from Figures 4a that the expansions of the very low C_3A content cements, namely cements A1, A2 and A3, were low; the highest expansion at 7 days, 40 micro-strain, was shown by cement A1 with an SO_3 content of 4.25 per cent. The 7 day expansions of all the remaining very low C_3A content cements were less than 20 micro-strain.

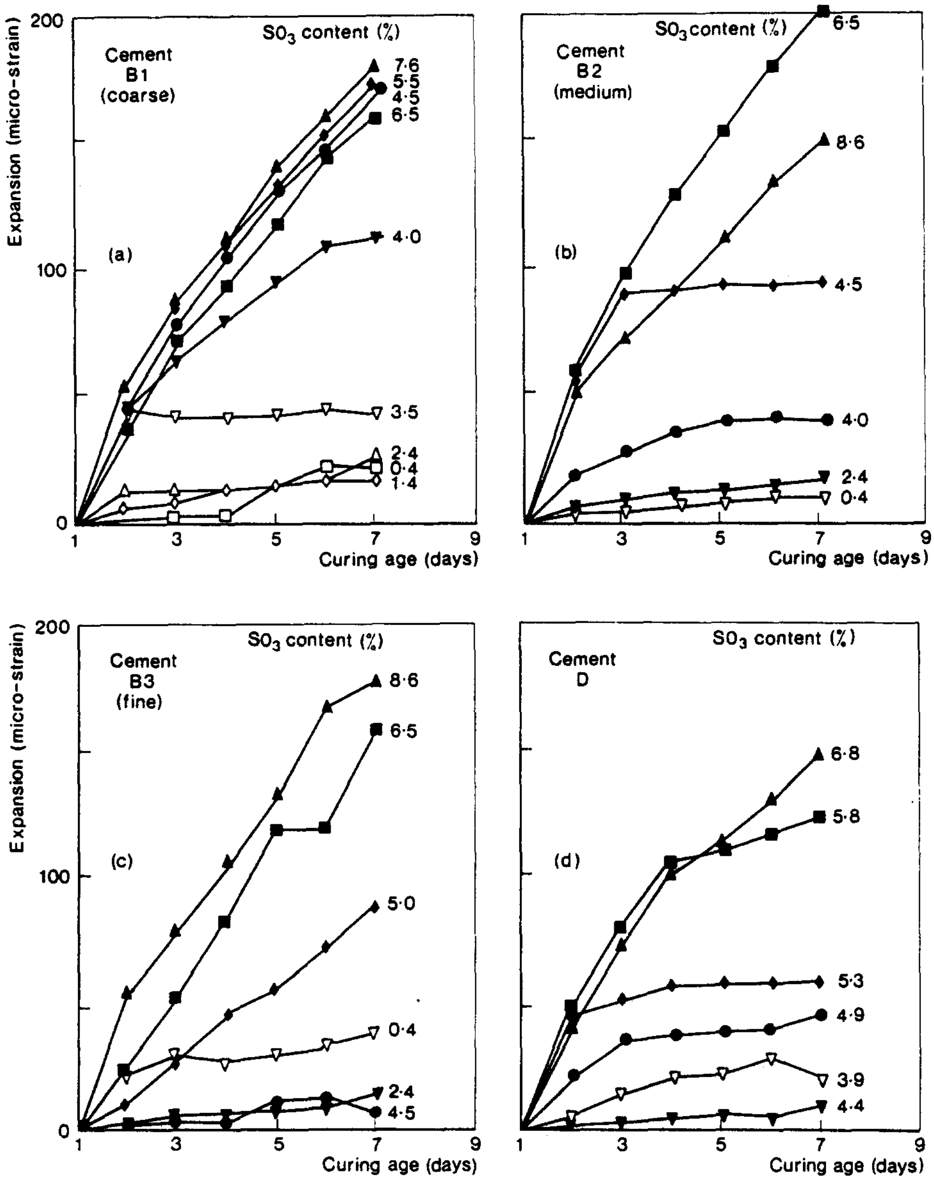


FIG. 3a - d

Variation of Expansion with Curing Age and SO_3 Content.

The generalised expansion/curing age behaviour of cements B1 to J is illustrated in Figure 5a. As a general rule, expansion goes up as the SO_3 level is increased. However, at low SO_3 contents the expansion, in a number of instances, at first decreased as the SO_3 content was increased, whilst at the higher SO_3 contents the expansions tended to follow a common expansion/curing age curve and this was followed by a fairly abrupt departure from the common curve when the expansion of the cements containing "intermediate SO_3 contents" apparently ceased. The curing age at which expansion apparently ceased may be assumed to correspond with the time at which all the sulphate in solution is consumed. The highest expansion observed at 7 days was 250 micro-strain (cement E).

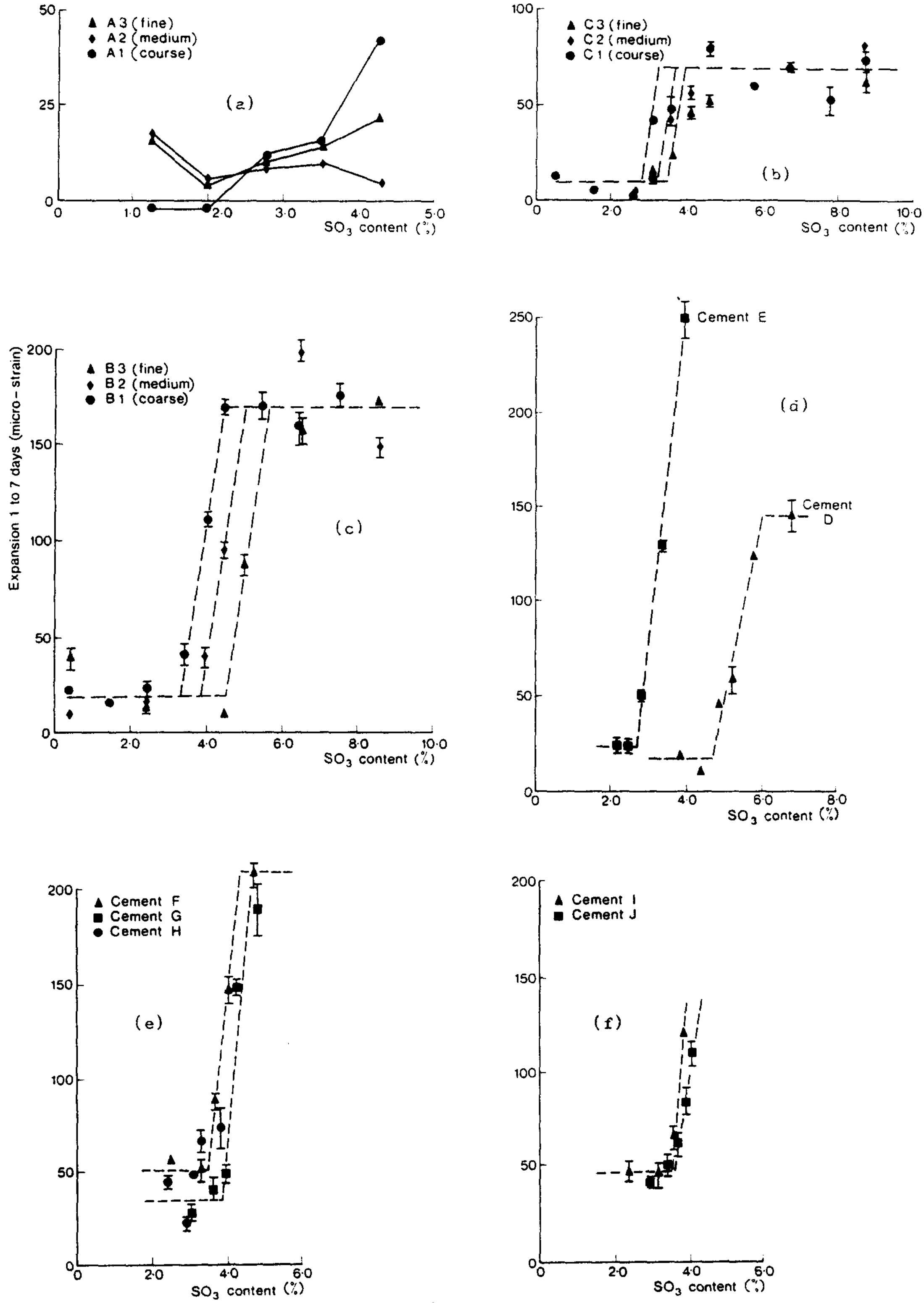


FIG. 4a - f

Variation of Expansion Occurring Between 1 and 7 Days and SO_3 Content.

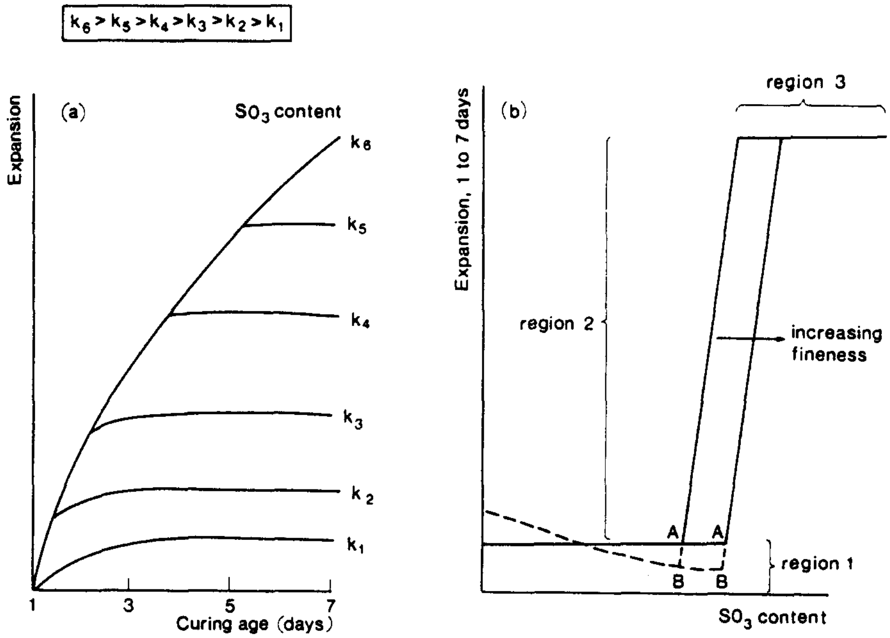


FIG. 5a-b

Expansion v Curing Age and SO_3 Content. Generalized Behaviour. Region 1 - 'Gypsum' Depleted in First 24 Hours. Region 2 - 'Gympsum' Depleted Between 1 and 7 Days. Region 3 - Excess 'Gypsum' Present After 7 Days.

The generalised 1 to 7 day expansion v SO_3 content behaviour of cements B1 to J is illustrated in Figure 5b. The total expansion occurring between 1 and 7 days was at first either constant and small or decreased as the SO_3 content was increased (see Figures 4b-4f). However, when the SO_3 content was raised above a particular level, which varied with cement fineness (see Figures 4b and 4c) and cement type, a sharp increase in 7 day expansion was observed, At still higher SO_3 contents the 7 day expansion was possibly independent of SO_3 content (see Figures 4b-d) and cement fineness (see Figure 4c).

In the small range of SO_3 contents within which sharp increases in 7 day expansion occur, the 7 day expansion decreases rapidly with increasing fineness (see Figure 4c) and is probably linearly related to SO_3 content (see Figures 4b-f), thus

$$S_{exp} = k_1 \ (SO_3\,\%) + k_2 \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

where S_{exp} is the expansion occurring between 1 and 7 days and k_1 and k_2 are constants. The measured slope k_1 ranges from 80 micro-strain/SO_3% for cements D and J to 170 micro-strain for cement E, the average slope being 130 micro-strain/SO_3%. The slope k_1 is probably independent of cement fineness but may be a function of the C_3A and 'C_4AF' contents, for example the ratio $C_3A/(C_3A + 0.75\ 'C_4AF')$ is lowest for cements D and J and is highest for cement E.

Thus to produce an expansion of 600 micro-strain in a Type S cement mortar it is necessary for between 4 and 7 per cent by weight of SO_3 to be consumed within the steep linear region. Therefore to compensate for shrinkage, a Type S cement should have an SO_3 content of between about 7 and 11 per cent, the actual amount required increasing with cement fineness and possibly also increasing with the ratio $(C_3A + 0.75\ 'C_4AF')/C_3A$. A point to note here is that Klieger and Greening (6) observed 7 day expansions in excess of 1000 micro-strain using a Type S expansive with an SO_3 content of only 4.5 per cent. It is suggested that this conflicting result may be due, in part, to the relatively high magnesium oxide content (3.0%) of their clinker (6).

From figures similar to Figures 3a to 3d it is possible to determine graphically the SO_3 contents below which all the gypsum in solution is consumed at 2,3,4,5 and 6 and 7 days curing. To do this we proceed as follows - we first note the expansions occurring in specimens with excess gypsum at 2,3,4,5,6 and 7 days. We then determine graphically from the 7 day expansion v SO_3 content plot the SO_3 contents that give similar expansions at 7 days.

As an example let us consider cement B1. From Figure 3a the expansion in specimens containing excess gypsum is approximately 50 micro-strain at 2 days and 80 micro-strain at 3 days. The SO_3 contents associated with similar expansions at 7 days are from Figure 4c, 3.6 and 3.75 per cent respectively. Continuing this procedure we deduce that the expansion v curing age behaviour of specimens of cement B1, containing SO_3 -contents below which all the sulphate in solution is consumed after 2 to 7 days curing is given by Figure 6.

The SO_3 contents below which all the sulphate in solution is consumed after 2 to 7 days curing are given in Table 2 for each of the cements tested. Also given in this table, in brackets, are the expansions corresponding with each quoted SO_3 content level.

Shrinkage

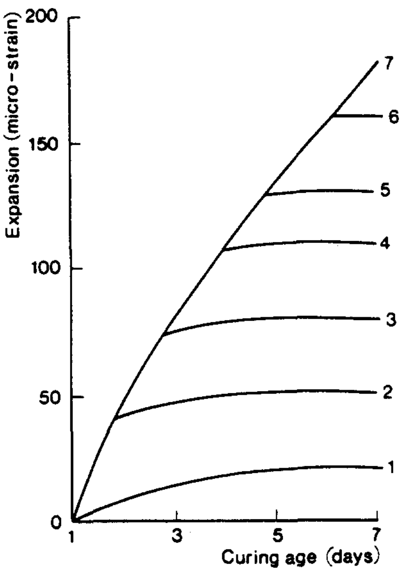
The mean drying shrinkage of the mortar specimens made from cements B2, D,E,F and G with a normal SO_3 content and with an SO_3 content such that all

TABLE 2

SO_3 Contents Below which all the Sulphate in Solution is Consumed After 2 to 7 Days Curing and the Corresponding Expansions+

Cement	SO_3 Content (%) [Expansion (micro-strain)]					
	Curing Age (days)					
	2	3	4	5	6	7
B1	3.60 (50)	3.75 (80)	3.90 (110)	4.05 (130)	4.20 (160)	4.30 (180)
B2	4.10 (50)	4.35 (80)	4.55 (110)	4.70 (130)	4.90 (160)	5.00 (180)
B3	4.80 (50)	5.00 (80)	5.15 (110)	5.25 (130)	5.40 (160)	5.60 (180)
C1	2.90 (20)	3.00 (40)	3.10 (50)	3.15 (60)	3.20 (70)	3.30 (80)
C2	3.40 (20)	3.50 (40)	3.60 (50)	3.70 (60)	3.75 (70)	3.80 (80)
C3	3.60 (20)	3.75 (40)	3.80 (50)	3.90 (60)	4.00 (70)	4.05 (80)
D	4.95 (40)	5.15 (80)	5.30 (100)	5.35 (115)	5.40 (130)	5.50 (145)
E	2.90 (60)	3.20 (110)	3.45 (160)	3.65 (190)	3.80 (220)	4.00 (250)
F	3.55 (60)	3.75 (100)	3.90 (130)	4.05 (160)	4.15 (185)	4.30 (210)
G	4.05 (70)	4.20 (110)	4.25 (120)	4.35 (140)	4.40 (160)	4.50 (180)
H	3.40 (60)	3.90 (75)				
I	3.70 (90)					
J	3.60 (55)	3.80 (80)	4.00 (95)	4.10 (105)		

+ The information in the Table is incomplete because in the case of cements H, I and J the gypsum in solution was depleted in the first few days of curing.



Curve	SO_3 content (%)
7	≥ 4·30
6	4·20
5	4·05
4	3·90
3	3·75
2	3·60
1	≤ 3·35

FIG. 6
Idealized Behaviour of Cement B1. Expansion v Curing Age, (The Curve Numbers are the Ages in Days at which the Sulphate in Solution is Consumed).

the sulphate in solution was consumed between a curing age of 4 and 7 days is shown plotted against drying period in Figure 7a to c. With each of the latter 'cements' the observed expansion at 7 days was greater than 100 micro-strain. Examination of Figure 7a-c shows that the drying shrinkage behaviour of the mortars is essentially the same at both the normal and the high SO_3 content levels. Similar drying shrinkage behaviour was observed at intermediate SO_3 content levels but greater drying shrinkage was observed at still higher SO_3 content levels.

The net shrinkage of the specimens at an age of 35 days is given in Table 3. The figures quoted are the sum of the expansion occurring between 1 and 7 days and the drying shrinkage occurring between 7 and 35 days. It is apparent from the Table that high SO_3 contents can provide some shrinkage compensation in a number of the cements tested, namely cements B1, B2, B3, E, F and G, but not with cements A, C, D, and possibly J. No shrinkage compensation was observed in the case of cements H and I probably because the gypsum contents employed were too low. Cement A is a sulphate resisting cement and of the remaining cements C, D and J had the lowest C_3A contents determined by both the Bogue compound composition and the X-ray diffraction analyses. Little shrinkage compensation is therefore to be expected with these cements.

Conclusions

Expansion and shrinkage measurements were made on a number of cements with SO_3 contents in excess of the limits specified in BS 12. Although the following conclusions are only strictly applicable to the cement mortars tested it is nevertheless suggested that a number of the conclusions may be generally applicable to oversulphated Portland cements and Type S expansives.

1. With the exception of the very low C_3A content cement a sharp increase in 7 day expansion is generally observed when the SO_3 content of the cements is raised above a particular level which varies with cement fineness and

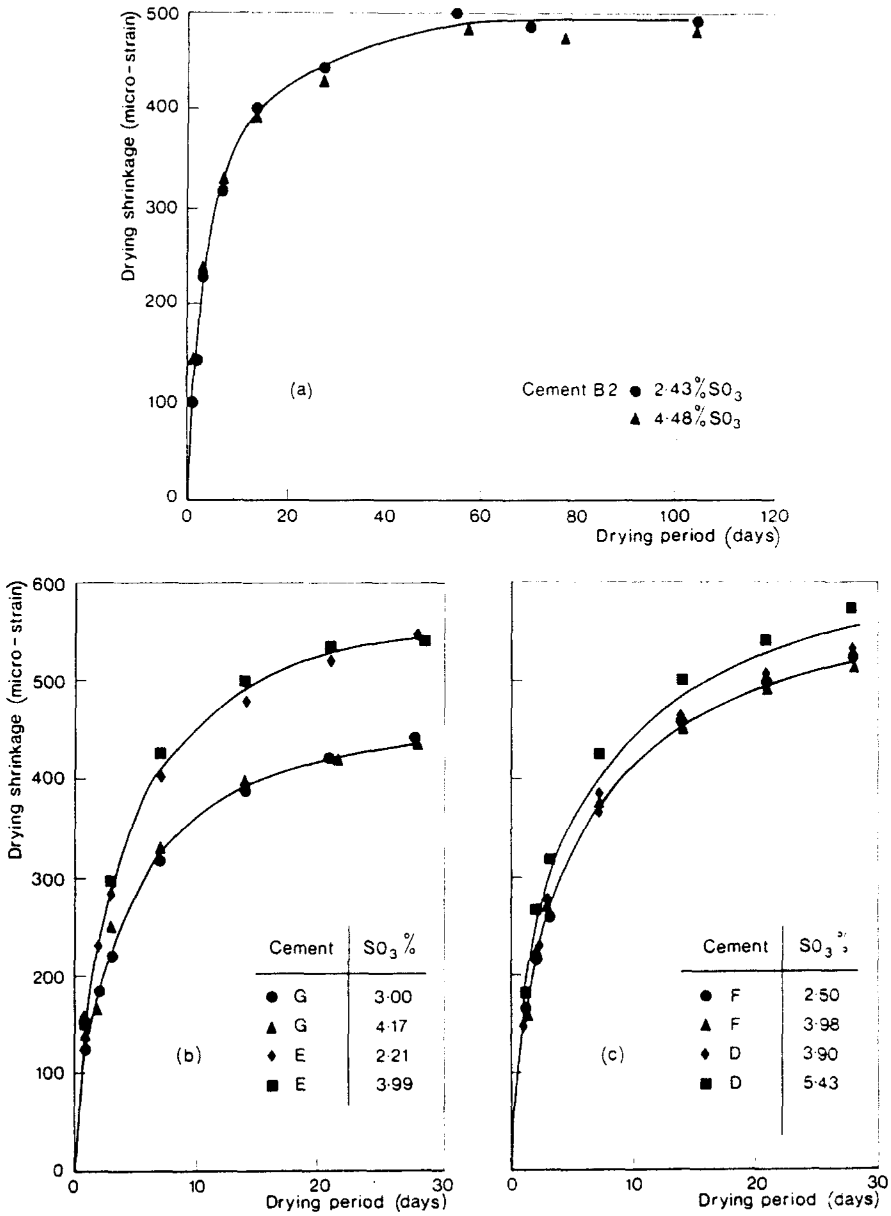


FIG. 7a-c

Mean Drying Shrinkage of Mortar Specimens with a Normal and a 'High' SO_3 Content.

composition. At still higher SO_3 contents the 7 day expansion is probably independent of SO_3 content.

2. The largest observed expansion at 7 days was 250 micro-strain.

3. In the small range of SO_3 contents within which sharp increases in 7 day expansion occur, the 7 day expansion is linearly related to SO_3 content thus:

$$S_{exp} = k_1 \; (SO_3\,\%) + k_2$$

where S_{exp} is the expansion that occurs between 1 and 7 days water curing and k_1 and k_2 are constants.

TABLE 3

Mean Net Shrinkage at an Age of 35 Days

Cement	Mean Net Shrinkage (micro-strain)										
	SO_3 Content (%)										
	0.42	1.19	1.95	2.72	3.49	4.25					
A1	-	468	368	348	429	457					
A2	-	482	379	361	435	-					
A3	-	519	416	402	410	458					
	0.38	1.41	2.43	3.46	3.97	4.48	5.00	5.51	6.53	7.56	8.58
B1	553	478	391	373	333	318$^+$	-	393	381	377	-
B2	474	-	423	-	391	329	-$^+$	-	369	-	386
B3	516	-	431	-	-	411	412	-$^+$	362	-	350
	0.51	1.53	2.56	3.07	3.58	4.09	4.60	5.63	6.65	7.67	8.69
C1	463	421	360	379$^+$	399	-	391	425	415	428	409
C2	-	-	410	385	364$^+$	410	-	-	-	-	422
C3	-	-	-	401	447	455$^+$	449	-	-	-	533
	3.90	4.38	4.86	5.34	5.82	6.77					
D	512	547	476	517$^+$	437	524					
	2.21	2.40	2.80	3.40	3.99						
E	521	537	445	337	297$^+$						
	2.50	3.24	3.61	3.98	4.72						
F	469	455	412	363$^+$	361						
	3.00	3.58	3.88	4.17	4.76						
G	413	400	397	286$^+$	338						
	2.42	2.93	3.16	3.41	3.90						
H	370	399	411	373	393						
	2.36	3.10	3.55	3.84							
I	395	388	384	331							
	2.91	3.41	3.65	3.90							
J	347	338	324	318							

+ At higher SO_3 contents the gypsum in solution was not depleted at 7 days.

4. The measured slope, k_1, ranged from 80 micro-strain/SO_3% to 170 micro-strain/SO_3%.

5. The slope k_1 is independent of cement fineness.

6. In the range of SO_3 contents within which sharp increases in 7 day expansion occur the subsequent drying shrinkage is essentially independent of SO_3 content.

7. Significant shrinkage compensation is only achieved with the higher C_3A content cements.

8. It is suggested that the linear relationship between S_{exp} and SO_3 content may be used to determine from a limited number of 7 day expansion tests on a Type S cement, the SO_3 content below which excessive expansions do not occur after 7 days curing.

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References

1. D.W.Hobbs, To be published.

2. ACI Committee 223. Expansive cement concretes - Present state of the knowledge, J ACI Proceedings, 67, 583 (1970).

3. P.Klieger, NRMCA Publication, 134, 3 (1971).

4. A.H.Gustaferro, N.Greening and P.Klieger, J PCA Labs, 8, 10 (1966).

5. Working committee on sulfate resistance, ASTM Committee C1, ASTM Bulletin, 212, 37 (1956).

6. P.Klieger and N.R.Greening, Proceedings of the fifth International Symposium on the Chemistry of Cement, Tokyo. Part IV Admixtures and Special Cements. Supplementary Paper IV-132, 439 (1968).

7. D.W.Hobbs and A.R.Mears, Magazine of Concrete Research, 23, 89 (1971).

8. A.R.Mears, RILEM/CEMBUREAU International Colloquium on the Shrinkage of Hydraulic Concretes, Instituto Eduardo Torroja, Madrid, 1, paper 1E (1968).

9. A.R.Mears, Technical Report TRA 449, Cement and Concrete Association, London (1970).