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Reactor Experimental Engineering Division

SUMMARY REPORT ON PORTLAND CEMENT CONCRETES FOR SHIELDING

R. B. Gallaher
A. S. Kitzes

Final Report of Development Program on Concretes for Shielding Terminated March, 1951

Date Issued: FEB 7 1953

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ABSTRACT

Portland cement concretes of medium density have been developed, using a variety of aggregates to impart specific nuclear properties to the finished product. Concretes containing 1% boron as colemanite and 8% water are described, and physical property data for various concretes are given.

Thermal conductivity for barytes concrete was found to be:

Temperature (^{o}F)	122	212	257	302	392	482
k (Btu/hr-ft-^{o}F)	0.926	0.997	1.02	0.872	0.866	0.745

The specific heat (Btu/lb-^{o}F) of barytes concrete increased linearly with temperature from 0.123 at 122° F to 0.150 at 392° F.

INTRODUCTION

The experimental program conducted at ORNL on concretes suitable for pile shielding has been concerned with the development of highly hydrated concretes with acceptable mechanical properties. Early work at the Laboratory was concerned with concretes containing magnesium oxychloride cement, which has a high fixed water content, but when further study revealed that the aging characteristics of MO concrete (magnesium oxychloride cement, iron punchings, and steel shot) were poor[1], interest turned toward the possibility of formulating portland cement concretes possessing characteristics suitable for pile shielding.

Among the properties desired in a concrete for a reactor shield are:

1) High density to minimize thickness

2) High hydrogen content (water) to thermalize intermediate neutrons

3) High content of heavy elements for degradation of fast neutrons as well as gamma rays

4) Low cost of ingredients

5) Ease of mixing and placing the concrete

In addition, structural strength, stability under radiation and stability under hot moist or dry conditions.

The specific use of the shield will determine which of the above requirements are important. For example, the size or weight of the shield is less important in a stationary reactor than in a mobile reactor, but the total cost of the shield may be more important.

Also, provision for neutron moderation and capture is important in a reactor shield but is of no importance where the shielding is entirely against gamma rays or x-rays. Cheap shielding, other than ordinary concrete, precludes the use of lead, tungsten and other heavy metals as the aggregate for concrete and limits one to the use of various iron ores, scrap iron and barytes. The high density requirement for concrete shields limits the use of low density additives such as boron and water containing compounds. It is rather difficult to formulate a concrete with a high hydrogen content and at the same time, try to meet a high density specification without using expensive aggregates.

This report summarizes the results of work at ORNL to determine those properties of special portland cement concretes which must be considered in designing reactor shields. Results of physical tests, induced radiation, decay curves, composition and cost data are presented.

HEAVY AGGREGATE CONCRETE

Barytes Concrete

The history and development of barytes concrete, which is similar to ordinary portland cement concrete except for density, have been adequately discussed by Tirpak[2]. In order to obtain the extra weight in barytes concrete (sp. g. = 3.5), the sand and gravel commonly used in ordinary concrete (sp. g = 2.3) were replaced by barytes aggregate ($BaSO_4$). The handling and pouring of barytes concrete were similar to that of ordinary concrete except for the fact that the extra weight of the barytes would often stall a regular mixer when filled to capacity. For this reason, allowance was made for the 50% additional weight when loading the mixer.

Aggregate - Barytes, which contains at least 90% $BaSO_4$ for the better grades, was obtained at a cost of $18 per ton, delivered, from L. A. Woods and Sons, Sweetwater, Tennessee. Table I gives the chemical composition and gradations of the Sweetwater aggregates. The difference in iron content of the coarse and fine aggregate can be attributed to the concentration of Fe_2O_3 in the fines when the barytes was crushed. The larger barytes chunks are very friable and ordinarily would not be considered

TABLE I

PARTIAL ANALYSIS OF BARYTES AGGREGATE

Chemical Composition

	Coarse	Fine
$BaSO_4$	95.9	81.6
Fe	1.0	9.8
Ca	.5	.9
O_2	1.3	6.2

Gradation

Coarse Sieve Size	Coarse % Passing	Fine Sieve Size	Fine % Passing
1"	100	3/8"	100
3/4"	90-100	No. 4	95-100
3/8"	20-55	No. 16	45-80
No. 4	0-10	No. 50	10-30
		No. 100	2-10

Weight per cu. ft.:	162 lb Dry Rodded	159
Specific Gravity:	4.2	4.0
Voids	38%	35%

for use in concrete; however, the fine aggregate has excellent strength, and a satisfactory concrete resulted when the two were combined with cement.

In addition to the common impurities associated with barytes, opal and chalcedony, which have a high silicon content, may be present. Since opal and chalcedony react deleteriously with high alkali cements, all barytes aggregate should be chemically tested[3] for alkali reactivity to determine whether or not these impurities are present in sufficient quantities to ultimately cause the concrete to fail due to expansion cracks. Aggregates containing more than 0.24 percent by weight of opal and 5 percent of chalcedony are recommended for use only in cements of low alkaline content.

Barytes obtained from the western part of the United States was used in a series of experiments; however, results of a petrographic analysis and chemical tests by the Bureau of Reclamation at Denver, Colorado[4], showed that alkali-reactive opal and chalcedony were present in sufficient quantities to warrant discarding the entire shipment.

Formulation and Properties - The formulation and physical property data shown in Table II were determined using Sweetwater barytes. Procedures and equipment used to determine these physical properties are reported in Appendix B. Thermal conductivity and specific heat data are also included although these two properties are not important in ordinary concrete work. These properties, however, must be considered in reactor shield design because the energy of the various radiations escaping from a reactor is converted into heat when absorbed in the shield. This heat,

TABLE II

FORMULATION AND PHYSICAL PROPERTY DATA

A. Formulation for a barytes concrete with a density of 3.5 (5900 lb/yd^3)

1) For pouring

	Weight % (wet)	Weight % (dry)	Pounds per yd^3 mix
Sweetwater Barytes (1")	45.15	48.1	2660
Sweetwater Barytes (3/8")	39.23	41.9	2320
Portland Cement (Type I or II)	9.37	10.0	550
Water	6.25		370 (44.5 gal)

2) For blocks

	Weight % (wet)	Weight % (dry)	Pounds per yd^3 mix
Sweetwater Barytes (1/2")	87.9	91.4	5270
Portland Cement (Type I or II)	8.3	8.6	500
Water	3.8		230 (27.5 gal)

B. Physical Property Data

1) Compressive Strength

Age(days)	Compressive Strength (psi)*	Curing in Wet Sand (days)
8	2897	5
28	3607	27
57	3977	27
85	4153	27
91	4453	27
112	4207	27

* Average of three 6" x 12" cylinders

TABLE II (CONTINUED)

2) Shear Strength - 845 psi

3) Thermal Conductivity (Figure 2)

Temperature (^{o}F)	122	212	257	302	392	482
k(Btu/hr-ft^2-^{o}F/ft)	.926	.997	1.02	.872	.866	.745

4) Specific Heat

Temperature (^{o}F)	122	212	302	392
c_p	.123	.133	.141	.150

5) Linear Expansion or Contraction on Setting at 100°F

$+ 8 \times 10^{-4}$ in/in after 2-3 days

$+ 5 \times 10^{-4}$ in/in after 28 days

6) Water Content Upon Being Submerged in Water

Time (days)	0	1	7	14	28
Water Content (percent)	5.61	5.07	5.59	5.54	5.69

7) Pile Induced Radiations (ORNL-973 p.31)
Figure 1

8) Cost

The cost of barytes concrete is higher than for ordinary concrete because of the aggregate; handling and pouring charges are also higher because of the additional weight of the concrete.

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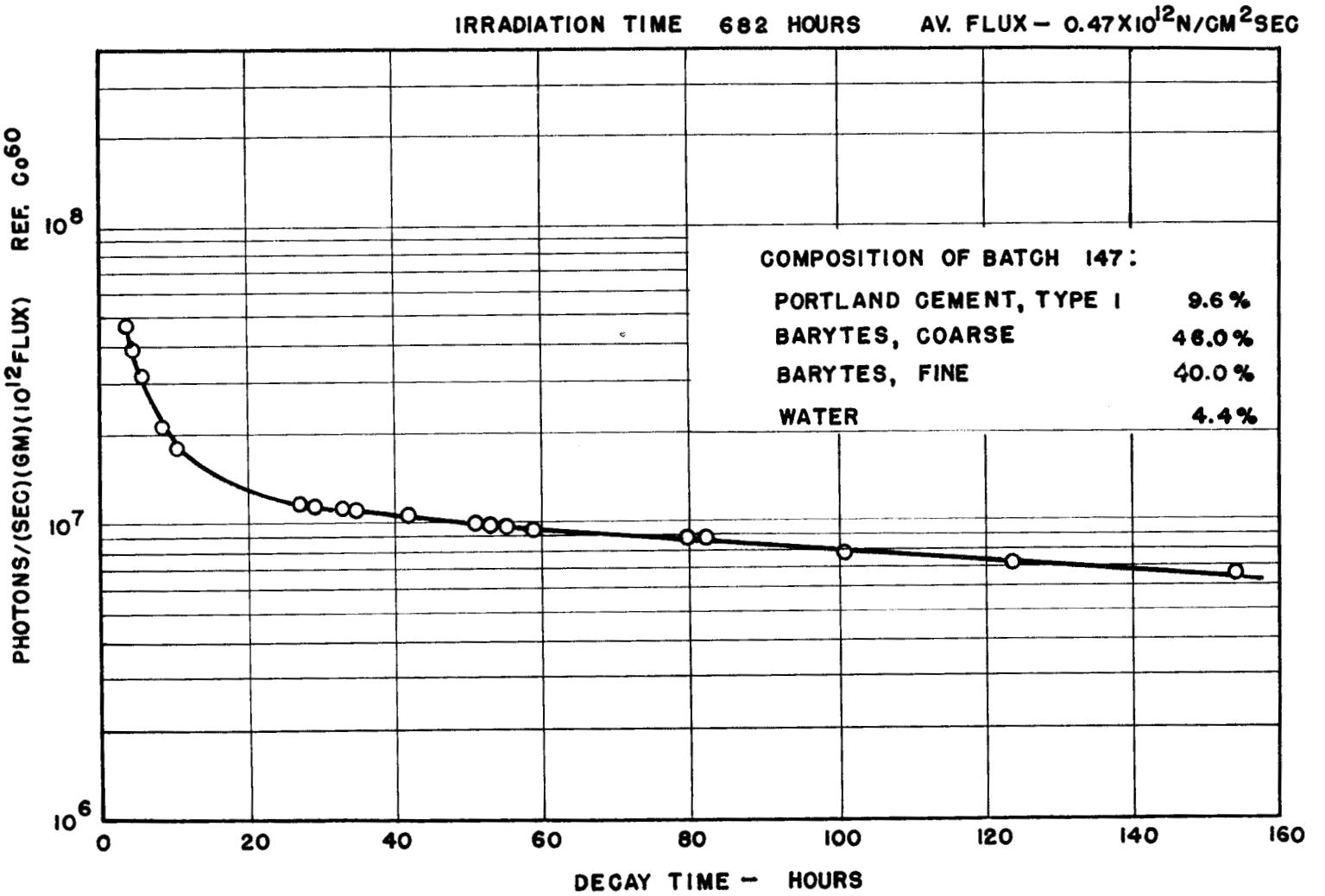

FIG. 1. SHORT TIME GAMMA DECAY OF BARYTES CONCRETE (BATCH 147)

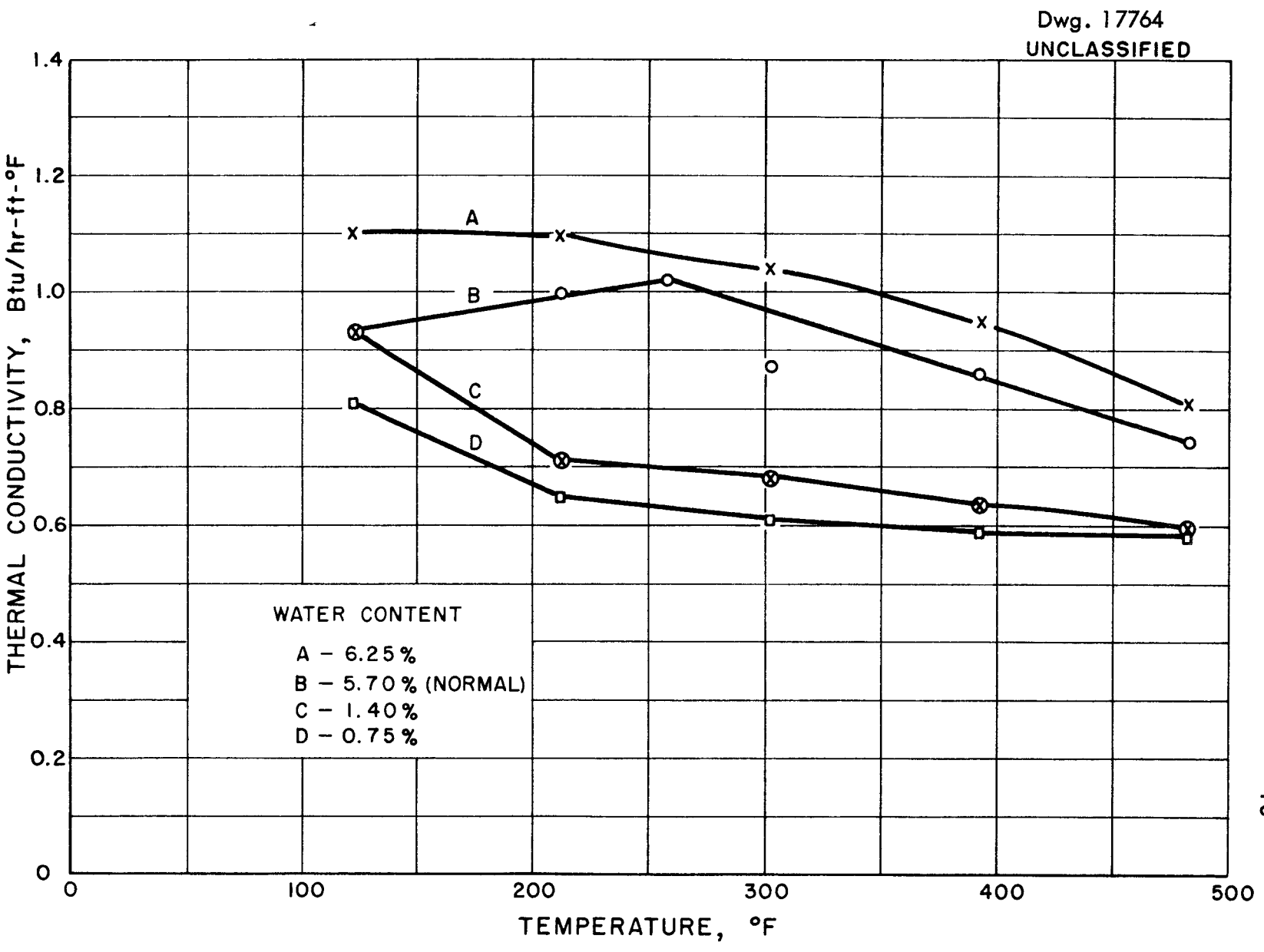

Fig. 2 Thermal Conductivity-Temperature Relationship of Barytes Concrete of Various Water Contents.

if not removed rapidly, could create excessive thermal stresses in the concrete causing failure or could cause loss of water by evaporation if the local temperature of the concrete approached the boiling point of water.

Comparison of Barytes and Ordinary Concrete - The physical properties of barytes concrete compare very favorably with those of ordinary concrete (Table III). Concretes of comparable compressive strengths can be formulated using either barytes or limestone aggregates. The thermal conductivity at 200°F for both concretes are the same; however, the specific heat of the barytes concrete is 20% lower at room temperature than that for ordinary concrete.

TABLE III

COMPARISON OF BARYTES AND ORDINARY CONCRETE AT ROOM TEMPERATURE

	Barytes	Ordinary
Density, g/cm^3	3.5	2.3
Density, lb/ft^3	218	144
Compression Strength after 28 days curing, psi	3500	3500
Thermal Conductivity Btu/hr.ft.°F at 200°F	1.0	1.0
Specific heat, Btu/lb.°F	0.123	0.156
Cost, $\$/yd^3$ (materials only)	~ 55	~ 6.50

Iron Portland Concrete

A very dense Portland cement concrete using iron aggregate was developed to replace MO concrete. The concrete had good workability and high density (~ 6) but a low water content (approximately 3%).

Formulation - Properties - The amounts of each ingredient actually used are shown in Table IV.

TABLE IV

HIGH DENSITY PORTLAND CONCRETE

	Pounds/yd^3	Weight Percent (dry basis)
Iron Punchings	5070	51.9
1110 Shot	2280	23.4
330 Shot	1520	15.6
Portland Cement (Type I)	890	9.1

The above-described mix was calculated to give a density of 5.8 gms/cm^3, but the test cylinders, which were steam cured for 7 days, had a density of approximately 6.0 due to "heeling" in the mixer--the fine aggregate and cement phase sticking to the walls of the mixer.

Aggregate - Iron punchings (slugs) ranging from 1/4 inch to 1 inch in diameter and thickness and two sizes of chilled shot were the aggregate used in this concrete. SAE 1110 and SAE 330 shot were 1/8 inch and 1/32 inch in diameter, respectively. The layer of rust on the punching was not removed prior to use.

Physical Properties - Table V gives the compressive strength of this high density concrete.

TABLE V

COMPRESSIVE STRENGTH OF HIGH DENSITY CONCRETE USING PORTLAND CEMENT AS THE CEMENT PHASE

Age (days)	Compressive Strength (psi)*	Curing Time (days)
8	3319	6
32	3663	27
91	5288	27

*Average of 3 cylinders

Handling and Pouring Heavy Aggregate Concrete - Each step in handling and placing the concrete should be carefully controlled to maintain uniformity within the batch and from batch to batch so that the completed shield has uniform quality throughout. It is essential to avoid separation of the coarse aggregate from the mortar or of water from the other ingredients. For these reasons, it is recommended that heavy aggregate (iron) concrete be placed in large sections by the internal vibrations technique. The fine aggregate (steel shot) and cement phase are mixed in an ordinary mixer and placed in the forms. The steel punchings are then added with heavy continuous internal vibration. Iron densities up to 70-80% by volume can be achieved in this way.

The internal vibration technique for placing heavy aggregate concrete has several advantages.

1) Eliminates "heeling" and assures uniform maximum density in the forms

2) Accepted batching techniques for regular concrete can be used

CONCRETES WITH HIGH WATER CONTENTS

Barytes-Limonite Concrete

By adding limonite ($2Fe_2O_3 \cdot 2H_2O$) to the mix the water content of barytes concrete was increased from 5.6% to 13%; however, this decreased the density from 3.5 to 3.2.

Formulation - Approximately 300 blocks, 2' x 2' x 1', were cast commercially, using the mix shown in Table VI to give a 3,000 psi concrete.

TABLE VI

FORMULATION FOR BARYTES-LIMONITE CONCRETE

	Pounds/yd^3	Weight Percent
Barytes Coarse (3/4")	2210 lbs.	40.4
Barytes Fine (3/8")	1050 lbs.	19.2
Limonite (crushed)	1220 lbs.	22.3
Portland Cement (Type I)	610 lbs.	11.1
Water	46 gals.	7.0

Physical Properties and Handling - Test cylinders 6" x 12", poured at ORNL, were found to have a density of 3.25 and a seven-day compressive strength of 3750 psi. Test cylinders 6" x 12" poured during commercial casting had an average density of 3.17, ranging from 3.05 to 3.22, and the 14-day compressive strength of the field cylinders averaged about 3600 psi.

No difficulty was encountered in pouring the barytes-limonite mix, and twelve hours after pouring, the blocks had sufficient strength to support their own weight. Standard portland cement curing technique was used.

BORON CONTAINING CONCRETES

If a large fraction of the thermal neutrons emerging from a pile can be destroyed by (n, γ) capture, without producing hard gamma radiations, and if hydrogen is present in sufficient quantities to thermalize intermediate neutrons, the total hard gamma flux to be shielded against is reduced by several orders of magnitude. To accomplish this, concretes containing boron, high water contents, and densities greater than 3.0 were developed. Highly insoluble boron compounds and control of the gradation of the fine aggregate were used to minimize the detrimental effects of the boron additions on setting time. Materials with high water of crystallization were incorporated to increase the water content of the concretes.

Concrete with Colemanite as the Boron Containing Compound for Block Fabrication

Formulation and Aggregate - Even in small quantities, soluble boron retards the initial set and often prevents concrete from developing its full strength. To overcome this, calcium chloride additions were made, and up to 7.5 percent by weight of cement had little effect in counteracting the action of the boron, even when high early portland (Type III) was used with finely divided colemanite (100% through 200 mesh). Because of its large surface area the colemanite dust was sufficiently soluble to act harmfully on the cement.

A concrete containing 1% boron, 8% water, and a density of 3.1 was developed for the fabrication of blocks. For this mix, the aggregate and the boron containing material, Colemanite ($2CaO \cdot 3B_2O_3 \cdot 5H_2O$), were specially graded as shown in Table VII.

TABLE VII

ANALYSIS, GRADATIONS OF AGGREGATE AND FORMULATION OF MIX FOR BORON-CONTAINING CONCRETE

a) Formulation per Cubic Yard

	Pounds	Weight Percent
Barytes	3920	72.22
Colemanite	400	7.14
Portland Cement, Type I	830	14.96
Water	37.5 gal	5.6

b) Barytes - Gradation

Sieve Size	% Passing
1/2	100
No. 4	95-100
No. 16	45-80

c) Colemanite Gradation

Sieve Size	% Passing
No. 16	100
No. 50	10-30
No. 100	2-10

d) Colemanite Analysis

CaO	27%
B_2O_3	43-45%
H_2O	28-30%

Properties - ORNL test cylinders, after being cured in wet sand at room temperature for six days, had a density of 3.2 and a compressive strength of 2500 psi.

Curing Technique - When standard block-curing procedure (steam curing at 170° F for 24 hours) was used for colemanite blocks, they had no strength and crumbled at the end of the normal curing period. Apparently, the elevated temperature slightly increased the solubility of the colemanite, detrimentally affecting the initial set and early strength of the portland cement. The blocks were finally cured by cold water fogging for seven days.

Concrete Using Boron Frits

Formulation and Aggregate - A boron compound more insoluble than colemanite was found to prove satisfactory. A mix containing 1% boron, Type I portland, and barytes was developed, which resulted in a concrete with a density of 3.1. In this mix, boron frits, formed by fritting borax with silica, and obtained from the Chicago Vitreous Enamel Products Company, Cicero, Illinois, were used. The mix formulation is shown in Table VIII.

TABLE VIII

BORON CONCRETE USING FRITS

a)

Mix	Pounds/yd^3	Weight Percent
Barytes Coarse (3/4")	3120	56.8
Barytes Fine (3/8")	770	14.0
Frits	670	12.2
Portland Cement (Type I)	660	12.0
Water	280 (33 gal.)	5.0

b) Analysis of Frits

SiO_2	31.00	BaO	12.90
Al_2O_3	1.70	ZnO	6.70
B_2O_3	27.40	Na_2O	12.20
CaO	5.20	K_2O	1.00
		F	1.90

Physical Properties and Curing Technique - No physical property determinations were made. The mix can be handled and cured as barytes concrete.

Comparison of Concretes Using Colemanite or Frits as the Boron-Rich Material

Depending upon the shielding requirements, satisfactory concretes containing either colemanite or frits can be formulated. Concrete with frits would cost more because of the frits, contain less water for the same boron content but would be easier to formulate and cure. Concrete using colemanite, on the other hand, would be cheaper, contain more water, but would be more difficult to formulate because of the aggregate gradation specifications and would require special curing techniques.

CONCRETES USING LUMNITE AS THE CEMENT PHASE

Lumnite-Colemanite-Barytes Concrete

To take advantage of the water of crystallization of colemanite, other cement phases were investigated to find an alternate to portland cement for concretes containing this boron compound. Lumnite, a calcium aluminate hydraulic cement manufactured by the Universal Atlas Cement Company, New York, New York, was the first alternative tried.

Formulation - A successful mix of Lumnite, colemanite, and barytes (see Table IX) was formed, producing a concrete with a density of 3.1. The initial set of this concrete was retarded slightly.

TABLE IX

LUMNITE AND COLEMANITE CONCRETE

a)

Mix	Pounds/yd^3	Weight Percent
Barytes Coarse (3/4")	2990	57.2
Barytes Fine (3/8")	695	13.3
Colemanite	334	6.4
Lumnite	795	15.2
Water	413 (49.5 gal.)	7.9

b) Chemical Analysis of Lumnite

SiO_2	9.1
FeO	5.9
Fe_2O_3	4.7
$Al_2O_3 + TiO_2$	41.6
CaO	36.8
MgO	1.0
SO_3	0.17
Insoluble residue	0.70

Properties - After curing the test cylinders by water fogging for 20 hours, they had developed about 75% of their full strength ($\sim$ 3500 psi).

Curing Technique - Lumnite, like portland cement, sets and hardens when mixed with water. In less than 24 hours after being placed, it can be subjected to the full load for which it is designed. The curing techniques for ordinary portland cement concrete are not satisfactory for Lumnite, as it is necessary to spray the surface of the concrete with water within 24 hours after mixing. Because of this, the formulation given in Table IX was abandoned in favor of a mix containing Lumnite and portland cement, so that ordinary curing methods could be used.

Lumnite-Portland Cement-Colemanite-Barytes Concrete

Formulation - Portland cement is not usually added to Lumnite because it greatly accelerates the initial set, sometimes causing flash-set. However, the addition of partially soluble boron (colemanite dust) counterbalanced the effect of the portland cement, giving a product which could be handled within a reasonable length of time, before hardening was evidenced.

Thirty-six blocks, 2' x 2' x 1', were poured commercially, using the mix reported in Table X.

TABLE X

LUMNITE-PORTLAND CEMENT-COLEMANITE-BARYTES CONCRETE

	Pounds/yd^3	Weight Percent
Barytes Coarse (3/4")	3490	64.18
Barytes Fine (3/8")	210	3.87
Colemanite (dust)	350	6.45
Portland Cement (Type III)	460	8.5
Lumnite	460	8.5
Water	55 gals.	8.5

Properties - For this formulation the mixing time was held to a maximum of three minutes, the mixer being emptied within 30 minutes. The finished block, containing 8% water and 1% boron, had a density of 3.1. Twelve hours after pouring, the forms were removed and the blocks were lifted by anchor rings, which had been embedded during pouring. Standard curing procedures were used. Test cylinders of this mix had a one-day compressive strength of 1300 psi, and after curing for six days in wet sand the strength increased to 3000 psi.

ACKNOWLEDGEMENTS

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APPENDIX A

ADDITIONAL PHYSICAL PROPERTY DATA

Elemental composition of the various mixes discussed in this report are shown in Table XI and Table XII.

TABLE XI

CHEMICAL COMPOSITION - WEIGHT PERCENT

% Wt.	Barytes for Blocks	Hi-Density	Barytes Lumonite	Barytes Colemanite	Barytes Boron-frits	Lumnite Colemanite	Lumnite-PC Colemanite
Fe	8.78	87.50	13.66	1.03	2.19	3.07	1.87
H	0.43	0.33	1.02	0.85	0.56	1.09	1.10
B				0.98	1.04	0.88	1.02
O	31.03	5.82	37.70	34.89	33.80	36.95	36.98
Mg	0.38	0.13	0.23	0.22	0.23	0.14	0.20
Ca	4.53	3.96	5.37	8.46	6.26	5.48	7.67
Ba	41.93		31.97	40.70	40.13	38.59	38.03
Na	.13		0.09	0.11	1.21	0.11	0.11
Si	1.74	0.91	1.35	1.76	3.31	0.96	1.49
Al	0.57	0.33	0.50	0.61	0.64	1.76	1.32
Mn	0.07	0.35	0.14	0.01	0.02	0.12	0.04
S	9.94	0.05	7.58	9.63	9.15	9.06	8.97
Zn					0.66		
K					0.10		
F					0.23		
Ti						1.27	.071

TABLE XII

DENSITY (g/cm^3) ANALYSIS OF CONCRETE

g/cm^3	Barytes for Blocks	Hi-Density	Barytes Limonite	Barytes Colemanite	Barytes Boron-frits	Lumnite Colemanite	Lumnite-PC Colemanite
Fe	0.307	5.075	0.444	0.033	0.071	.095	.058
H	0.015	0.019	0.033	0.027	0.018	.034	.034
B				0.031	0.034	.027	.033
O	1.090	0.338	1.226	1.118	1.099	1.144	1.147
Mg	0.013	0.008	0.007	0.007	0.007	.004	.006
Ca	0.159	0.230	0.175	0.271	.204	.170	.238
Ba	1.470		1.039	1.304	1.307	1.187	1.180
Na	0.005		0.003	0.004	0.039	.003	.003
Si	0.061	0.053	0.044	0.056	0.101	.030	.046
Al	0.02	0.019	0.016	0.020	0.021	.055	.041
Mn	0.003	0.020	0.004		0.001	.004	.001
S	0.348	0.003	0.246	0.308	0.298	.281	.278
Zn					0.021		
K					0.003		
F					0.007		
Ti						.039	.022
Poured Density	3.50	5.8	3.25	3.2	3.25	3.1	3.1

APPENDIX B

METHODS AND PROCEDURES USED FOR DETERMINATION OF PHYSICAL PROPERTIES

1) Thermal Conductivity

Radial heat flow through a 6" x 12" specimen was measured by the temperature rise of a metered amount of water passing through a heat exchanger located at the center of the sample. The temperature drop across the sample was measured with thermocouples cast into the specimen a known distance apart. Radiation, conduction, and convection losses from the ends of the sample were minimized by employing an electrically heated guard ring system. Under these conditions the thermal conductivity was computed by means of the relationship:

$$k = \frac{Wc_p\ (\Delta T)w\ \ln r_2/r_1}{2\pi\ L(\Delta T)_s}$$

k = thermal conductivity, Btu/hr-ft-^oF

c_p = specific heat of water, Btu/lb-^oF

W = water rate, lb/hr

$(\Delta T)_w$ = temperature rise in water ^oF

L = length of sample, ft.

$(\Delta T)_s$ = temperature drop in sample, ^oF

r_2 = distance of outer thermocouple from center of sample, ft.

r_1 = distance of inner thermocouple from center of sample, ft.

2) Specific Heat

The specific heat of concrete was determined by direct calorimetric measurement. A weighed sample of concrete, heated to the desired temperature, was dropped into a measured amount of water and the temperature rise of the water was recorded when equilibrium was established.

3) Compressive strength ASTM C39-49

4) Shear Strength ASTM C78-49

5) Aggregate Gradation ASTM C33-46