SULFUR BASED BLOCKS: STATE OF THE ART

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الخلاصـة :

دلت البحوث أن خاصية اللصق المتأصله في ماده الكبريت يمكن استغلالها بفعالية في تطوير مواد بناء حديثه . لقد أدت البحوث على استعمالات الكبريت في التشييد إلى تطوير طوب كبريتي بدءاً بالطريقة المألوفة يتجهيز الخليط الساخن لإعداد خرسانة الكبريت . هذا وقد استخدمت طرق التصنيع عدة أساليب تقنيه جديده تتضمن تقديم التكوين والترشيح وقد ركز جزء من هذا العمل التطويري على استغلال المواد الخام المحلية في صناعة الطوب الكبريتي . كما تصف ورقه البحث هذه الوضع الصناعي الراهن للطوب الكبريتي وتشير إلى التطورات الحالية .

ABSTRACT

Research on sulfur has shown that its inherent binding property can be effectively utilized in development of new construction materials. Research on new uses of sulfur in construction has led to the development of sulfur-based construction blocks. Beginning with the first developmental work, in which a conventional hot-mix method of preparing sulfur-concrete was used, the methodology of construction has evolved several new techniques, which include pre-molding and infiltration methods. Part of this developmental work focussed on the utilization of various indigenous raw materials for block construction. This paper presents the state-of-the-art on the construction of sulfur-based blocks, highlighting current developments.

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INTRODUCTION

Research in the last decade on new uses for sulfur in construction has led to the development of construction blocks using sulfur as a binder. Early attempts concentrated on the conventional hot-mix method of preparing sulfur-concrete, in which a hot mixture of coarse and fine aggregates is blended with molten sulfur at 130-140°C and the resulting hot mixture is then molded into blocks. Experimental homes were constructed with such blocks to demonstrate this application and draw attention to this new construction material [1, 2]. Research in this area since then has focussed on the development of new techniques that could be employed to improve products by eliminating the disadvantages of the hot-mix method, and to utilize indigenous materials other than aggregates.

A study at the University of Washington developed sulfur-based clay blocks, introducing first the concept of a two-tier bonding mechanism [3, 4]. The blocks are made from kaolin-sand-sulfur mixture in which kaolin serves as an initial binder for the extruding process and sulfur acts as the primary binder for the blocks.

The anticipation that Saudi Arabia would become a major producer of surplus sulfur in the foreseeable future and the fact that good quality coarse aggregates are in short supply in many parts of the Kingdom have induced researchers to explore methods of possible utilization of sulfur as an indigenous material in construction. The on-going research at the University of Petroleum & Minerals on sulfur-based products has identified new techniques for construction of high-strength, durable sulfur-based blocks [5-8].

The aim of this paper is to present the state of the art of construction of sulfur-based blocks and to disseminate current research and developments. Advantages and disadvantages of various methods are also highlighted.

SULFUR AS A MATERIAL

Sulfur, an element having atomic number 16, exists as rhombic and monoclinic crystals. The allotropic transformation of sulfur during cooling from a molten state results in a change of crystalline form from monoclinic to orthorhombic below 95.5°C. The latter crystalline formation induces high

stresses in sulfur-based products. The molton sulfur is straw-yellow and transparent and remains in molten state in the temperature range of $130-140^{\circ}$ C without significant change in viscosity.

The early developmental work on sulfur-based products began with elemental sulfur [9]. The brittleness of sulfur products and the reversible phase transformation of elemental sulfur which caused durability problems in thermal cycling and alternate wet-dry conditions were identified as major shortcomings of elemental sulfur for use as a building material [10].

In order to impart some plasticity, and to control the reversibility of sulfur formation, elemental sulfur has been reacted with dicyclopentadiene (DCPD) and oligomers to produce what is often referred to as plasticized or modified sulfur. Tests have shown that modified sulfur enhances the material properties of sulfur concrete and improves durability [10, 11]. In recent years, work on sulfur-based materials has used modified sulfur.

SULFUR-BASED BLOCKS

Sulfur-based blocks can be categorized into three groups: conventional sulfur-concrete blocks, molded clay blocks and infiltrated blocks. The last two categories of blocks make use of materials that function as initial binders in the molding process.

Conventional Sulfur-Concrete Blocks

These blocks are made from a hot mixture of coarse and fine aggregate (or fine aggregate alone) and sulfur (mix temperature $130-140^{\circ}$ C). The usual process consists of preheating aggregates and then adding sulfur to produce a molten hot mixture which is poured in a mold, compacted, and then left to cool at room temperature. The rapid gain in strength which is related to heat loss permits demolding of blocks after only a short period of casting.

To enhance workability and reduce sulfur usage without a loss of strength, fines in the form of fly ash or crusher powder are added, the usual amount being in the range of 5-15 percent of the total weight. The basic properties and mix design of sulfur concrete (SC) made with coarse aggregates can be found in references [12] and [13]. SC blocks have high compressive strength in the order of 50-60 MPa, exhibit good durability in freeze-thaw and wet-dry cycles and also in aqueous and corrosive environments, provided that the aggregates are sound and suitable for use in sulfur concrete.

It should be emphasized here that certain types of local coarse aggregates, particularly the marginal limestone-type aggregates that are widely used in the Gulf region, suffer seriously from lack of durability in water [8, 14, 15]. While the exact mechanism of such instability is not explicitly understood as yet, it is believed that such a phenomenon is attributable to a great extent to thermal stress and the differential volumetric expansion and contraction of the aggregates and sulfur in contact with moisture. The short supply of good quality aggregates has prompted research on the use of sand and fines alone in making sulfur-sand (S-S) concrete. Tests have shown that S-S composites can be used as concrete material and particularly as blocks [5], as they retains all the favorable material properties of SC and show excellent durability in humid and aqueous environments. The mix design and properties of SC are covered in references [16] and [17].

Compared to SC blocks, S-S blocks require a larger quantity of sulfur for similar workability and show a reduction in strength. The demand for a larger quantity of sulfur is attributable to the increased surface area of an all-fine composition. Using proper gradation of sand and mix design, compressive strength in excess of 25 MPa can be attained through controlled casting [5, 8]. However, in view of the higher sulfur content (18–24%) and absence of coarse aggregate, S-S is susceptible to extensive creep [18].

The major disadvantages of the hot-mix method of construction of S-S or SC blocks are that very little time is available for casting, compacting and finishing, and that the rapid change in the ambient temperature of the blocks during cooling causes volumetric change which, if unaccounted and uncontrolled, would result in an uneven surface finish. This is attributable to a high coefficient of shrinkage and expansion and low thermal conductivity.

Molded Clay Blocks

In an attempt to develop clay blocks without the conventional firing at high temperatures and to offset the disadvantage of the hot-mix method of SC or S-S blocks, the University of Washington developed

clay-based blocks, introducing for the first time the concept of two-stage bonding [3, 4]. In this process, sand and powdered kaolin and sulfur are used as ingredients. Measured quantities of these three raw materials are mixed with water to produce a moist mixture, which is then precompressed in a mold at a high pressure to extrude blocks. The extruded blocks are heated in an oven to drive off the moisture. The clay content of the blocks provides the initial binding necessary to provide and maintain the shape of the blocks. Following this drying operation, the blocks are heated to about 155°C to melt the sulfur powder within the body of the blocks. After allowing the required time for melting the sulfur, the specimens are allowed to cool and harden at room temperature. No problems are encountered during the melting period due to possible sulfur drainage and settlement of blocks. References [3] and [4] give details of mix proportions, the casting procedures and the influence of various pertinent factors such as compaction pressure, and contents of clay, sulfur, and mix water on the properties of blocks.

The average compressive strength of such blocks with 10% clay and 22% sulfur is in the range of 10-18 MPa, depending upon the initial compaction pressure applied during molding and the gradation of the sand. Higher strength is achieved by using higher molding pressure. The optimum clay content from the viewpoint of maximum compressive strength is reported to be about 10%. Excessive clay content may lead to surface cracking during drying. Test data show that the blocks are durable in the presence of moisture, provided that the clay is a non-expansive type, and one of good quality having low permeability. The average water absorption, which depends upon the mix, is in the vicinity of 1.2% by weight for a three-day soaking period for specimens having 10% clay and 22% sulfur.

This process is now patented by the Sulphur Development Institute of Canada for commercial manufacturing and all information about this proprietary product can be obtained from this source¹.

This process requires good quality clay for stable blocks with no surface cracks. Expansive clays are unsuitable. The method is energy-intensive, as the drying and melting period demands high consumption of energy.

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Infiltrated Blocks

As the supply of good quality clay in the Kingdom is not apparently large, research at the University of Petroleum and Minerals focussed attention on the possible utilization of locally-available indigenous materials in the development of blocks. The research has succeeded in identifying feasible new methods of constructing sulfur-based blocks.

The process utilizes the technique of sulfur infiltration [19] to provide the primary bonding. In addition, it requires the use of cementitious material that provides the initial binding strength necessary for casting and handling. The materials that serve this purpose are identified as marl, gypsum, lime, and Portland cement.

The first attempt at the development of infiltrated blocks studied extensively the use of marl [5, 6]. Marl is a silty-sand or sandy-silt material found in large quantities in various parts of the Eastern Province of the Kingdom. When mixed with water and dried, marl exhibits weak binding strength, attributable to the presence of carbonates. This property has made marl attractive for use in highway construction work. In the development of sulfur-marl (S-M) blocks, the relatively weak binding strength of marl has been successfully utilized to develop good quality blocks.

The process essentially consists of mixing an appropriate proportion of sand and marl with water to produce a moist mixture. 20% marl is considered ample for good casting. The moist mixture is lightly pre-compressed in a mold to form a block which is then demolded and dried in an oven to remove moisture. Following this drying, the temperature of the block is raised to about $130-150^{\circ}$ C. It is then immersed in molten sulfur ($130-140^{\circ}$ C) for infiltration at atmospheric pressure. Allowing a short period of infiltration, the block is removed from the sulfur bath, wiped clean and then allowed to cool at room temperature. Figure 1 shows a flow chart depicting the essential steps involved in their construction process.

The gain in strength of S-M blocks is related to the sulfur loading which in turn depends on the period of infiltration. As shown in Figure 2, a 5–10 minute infiltration period would be ample to develop a relatively high strength, as the rate of sulfur loading is quite rapid at the early stage of infiltration. Figure 3 shows the influence of marl on the compressive strength considering an infiltration period of 5

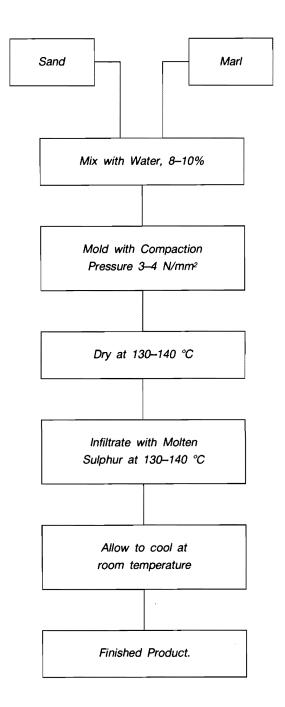
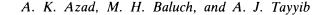


Figure 1. Flow Diagram for Preparation of S-M Blocks.

minute for all samples. As it is desirable to minimize the use of marl and to reduce the period of infiltration, an optimal mix proportion would be 20% marl and 80% sand for a five-minute infiltration period. It should be emphasized here that the infiltration period quoted is applicable to elemental sulfur. The use of modified sulfur would require slightly higher infiltration time for similar sulfur loading and development of compressive strength.



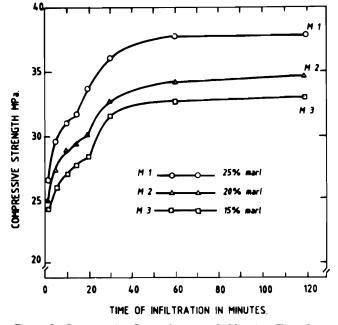


Figure 2. Compressive Strength versus Infiltration Time for S-M Specimens.

S-M blocks have excellent surface finish and good compressive strength (about 24 MPa with 5-minute infiltration) and are durable under alternate wet-dry cycling. In repeated wet-dry cyles over a period of two months, S-M blocks showed a 16% loss of strength without depicting any visual physical distress. The blocks have also a low permeability, as the water absorption, after 48 hours of soaking, is found to be less than 1.6% [5-6]. This absorption is, however, higher than the conventional SC blocks. S-M blocks are also slightly heavier than conventionally-dried clay blocks, with the unit weight of the former being about 2260 kg/m³.

The promising aspects of S-M blocks generated further research interest in utilizing materials other than marl, inasmuch as the quantity of marl is not as abundant as that of sand. Thus an effort was made to use only sand as filler material in the manufacture of infiltrated sulfur blocks. Cementitious materials, such as gypsum, lime, and cement were found to be acceptable as initial binders. The findings of the trial tests are given in the Fourth Progess Report of reference [8]. Of all these potential binders, the use of cement appeared to be most appealing and, therefore, a great deal of effort was mobilized towards the use of cement as an initial binder in the development of blocks.

The study of the influence of cement began initially with the modified S-M blocks in which a small quantity of cement was added to impart higher

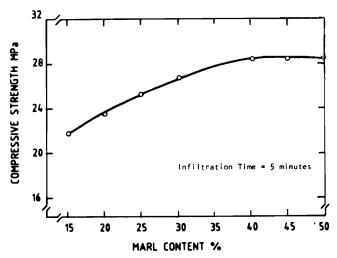


Figure 3. Compressive Strength versus Marl Content for S-M Blocks.

initial strength, which is desirable from the viewpoint of handling and effective retardation of sulfur loading during infiltration. The study [8] shows that the addition of about 4% cement partially fulfills these two objectives. The sulfur absorption drops by about 4% from the normal level for S-M blocks. This study has also shown that sulfur-based blocks can also be made from a mixture of sand and cement alone.

The development work at this stage envisioned the possible utilization of quarry dust as obtained from aggregate crusher plants. Large deposits of dust are seen at most crusher plants, particularly those located in the Eastern Province of Saudi Arabia, as the process of crushing poor marginal limestone aggregates produces considerable dust as a byproduct. Use of such man-made deposits as a raw material would not only be gainful consumption, but is also desirable from an environmental point of view.

Research at the University of Petroleum and Minerals shows that it is possible to make use of quarry dust in blocks [8, 20]. The raw materials for this process are sand, quarry dust, and cement. The casting procedure is essentially the same as for other infiltrated blocks. The moist mixture of sand, cement, and quarry dust is molded into blocks (S–D blocks) under a compaction pressure of 3.8 N/mm². After curing at room temperature for two days, the blocks are dried at $130-140^{\circ}$ C prior to sulfur infiltration. Figure 4 presents a flow diagram showing the essential steps involved in the preparation of S–D blocks.

The Sixth Progress Report in reference [8] gives

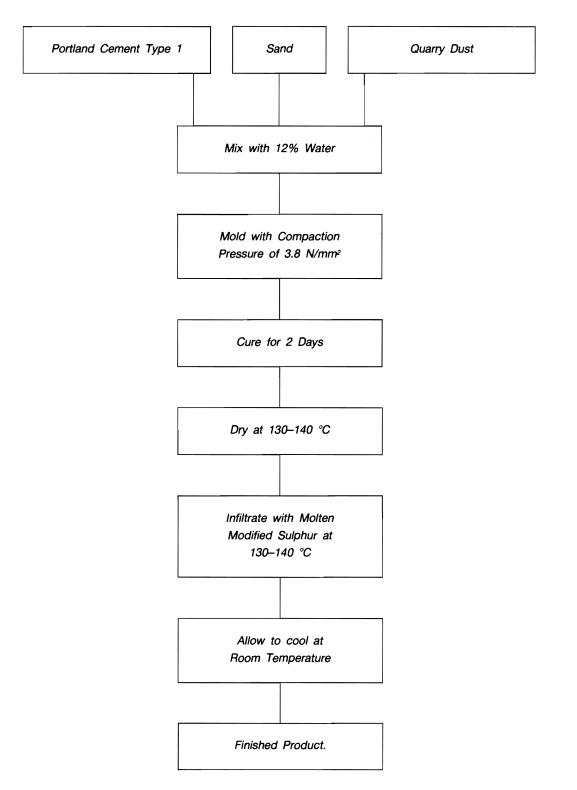


Figure 4. Flow Diagram for Preparation of S-D Blocks.

details of a parametric study of several variables such as the content of sand, dust, and Portland cement. While the cement content varied from 2% to 6% by weight, the sand to quarry-dust ratio was varied from 1:1 to 7:3. Results show that as the cement content increases, both sulfur loading and compressive strength decreases for a fixed period of infiltration. It is worthwhile to note that S-D specimens have a

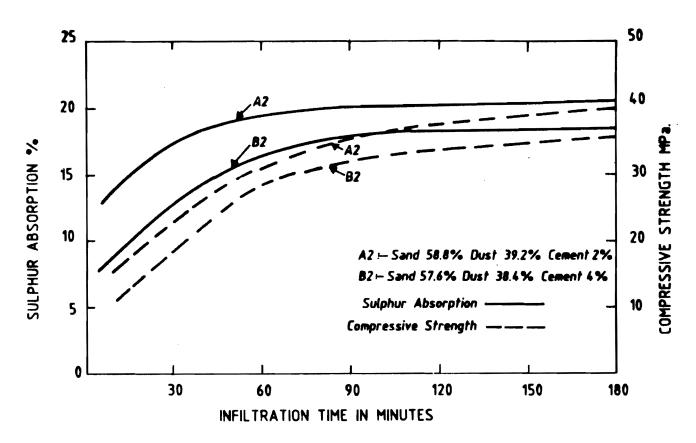


Figure 5. Variation of Sulfur Absorption and Compressive Strength with Infiltration Time.

considerably slower rate of infiltration than S-M blocks as depicted in Figure 5. The data in Figure 5 were obtained from the study of 50 mm (2 in) cube specimens. For S-D blocks, the satisfactory level of sulfur loading is achieved after about an hour of infiltration with modified sulfur. The time would be slightly less than this when elemental sulfur is used. Results have shown that the optimum mixture from the viewpoint of workability and strength is sand 67.2%, dust 28.8\%, and Portland cement 4%, representing a sand:dust ratio of 7:3. The water content is 12% by weight of dry mix. The resulting blocks have good compressive strength (27 MPa), durability, low permeability, and good surface finish like all other infiltrated blocks.

FUTURE WORK

The developmental work on sulfur-based blocks has convincingly indicated that good quality construction blocks can be produced using local raw material resources. However, for production at the industrial level and for adaptation to practical work, several pertinent factors need to be examined at depth. Reference [21] has listed several such items of importance which include, among others, laboratory work on identification of indigenous materials for use in blocks, fire rating and protection, jointing and plastering, and in-situ work on identification of habitat problems. The most pressing of all is the development of a suitable jointing (and plastering, if needed) technique for masonry work. It has been observed that conventional cement mortar cannot be effectively used with sulfur-based blocks. This is most likely due to chemical incompatibility and the possible reaction between sulfur and free lime in Portland cement mortar to form calcium polysulfide. Though mortarless construction is possible with interlocking-type blocks, it is essential to develop a simple jointing technique for sulfur based blocks, in order to facilitate their entry into the construction industry. In developing suitable mortar, the potential chemical reaction between sulfur and mortar should not be overlooked.

SUMMARY AND CONCLUSIONS

The developmental work on sulfur-based blocks has been reviewed by summarizing various methodologies of construction. As the construction of blocks is a promising area where sulfur can be gainfully utilized, the aim has been to disseminate current research to generate interest and draw attention to this potential new product.

The favorable material properties of sulfur-based blocks in terms of strength, physical appearance, durability, and permeability are compelling factors which warrant further research and development for practical use. It is envisaged that with a favorable sulfur market and proper research and development, sulfur-based blocks would become a viable commercial product gaining acceptance in many construction applications.

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