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MEMORANDUM

LITERATURE REVIEW

UTILIZATION OF ASBESTOS AND ASBESTOS-CEMENT PRODUCTS

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SUMMARY One phase of Project 191G55: "Modification of Cement" has been the addition of monomers (1) to cement mortar, with subsequent polymerization of the monomers by heat. Since the flexural and tensile strengths of the cement mortar are improved by this technique, a further step would be to see if this technique could improve the properties of asbestos-cement products, in particular to reduce the brittleness and increase the flexibility of sheet and pipe.

This literature survey was undertaken to gain background information for future laboratory work and also to disclose recent improvements to prevent duplication. Historical background, uses, chemistry and physical properties, and production methods of asbestos and asbestos-cement products are reported. Also reported are other Carbide projects involving asbestos.

No prior art bearing on the proposed experimental work was found.

INTRODUCTION Asbestos has gained wide acceptance in applications where heat or fire resistance or a strong fibrous binder is required. Major consumption of asbestos in the United States is in the manufacture of asbestos-cement sheet, pipe and other products. Interest in asbestos research has increased at Union Carbide since the acquisition of the Coalinga mine in California; very little work, however, has been done specifically with asbestos-cement.

Until very recently, a phase of the work of this group has been concerned with the polymerization of monomers in hydraulic cement systems. The project has had a degree of success; it is thought that a useful corollary would be the polymerization of monomers into asbestos-cement systems. A literature survey was therefore undertaken as a preliminary to such work.

Research and Development Department
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Commercial Importance of Asbestos

Asbestos was spun and woven into cloth as long ago as the first century AD (2). However, the steam engine, with its need for insulation and packing, stimulated great interest in the mineral so that its mining and usefulness were widely extended. Today, it is virtually indispensable to modern living.

Most of the asbestos consumed in the United States is used in the manufacture of building materials. In fact, asbestos-cement and asbestos-asphaltic product manufacture utilized 97 per cent of the total U. S. asbestos consumption of 726,000 short tons in 1962 (3). Other uses of asbestos, listed in the order of decreasing consumption, include abrasion-resistant frictional materials such as brake and clutch linings, flooring compositions, and textiles or felts (4). Fire-resistant sheet, roofing shingles, flat and corrugated siding, high pressure water pipe and sewer pipe comprise the bulk of the asbestos-cement products. These lists are, of course, incomplete; asbestos finds use in most any application where extreme heat resistance or strong fibrous binder is required.

Carbide Interest in Asbestos

In view of the unique properties of asbestos and importance of its products, much work has been done to discover more about its nature and technology. Interest in asbestos research has increased considerably at Union Carbide since the Nuclear Division acquired a Chrysotile (a type of asbestos) mine near Coalinga in Fresno County, California. The Division has subsequently constructed a processing plant at King's City, California (5). At present, several groups are engaged in various phases of research involving asbestos in general or with particular emphasis on Coalinga fiber.

A. Coalinga Fiber Research

At the Nuclear Division Research Center at Tuxedo in New York, Mr. G. S. Jordan and Mr. N. J. Setter are experimenting with reaction mixtures of Coalinga fiber and phosphoric acid. This material, designated as P-38, is reported to be compatible with several water soluble resins (particularly water soluble phenolics), resin latices and inorganic fibers. Moreover, they claim that sheets and pipe may be fabricated by extrusion to form tough and flexible products (6).

At the Technical Center in South Charleston, Dr. C. W. McGary's group is studying reactions of Coalinga asbestos with various compounds such as hydrochloric acid, maleic anhydride, alkylene oxides, and diisocyanates (7). Another project at the Technical Center under the supervision of Dr. J. A. Faucher and directed by Dr. G. D. Jacobs is concerned with improving the impact strength of inorganic polymers through the incorporation of tough organic polymers such as polycarbonates. The primary inorganic polymer studied in these experiments is the reaction product (P-38) of Coalinga fiber and phosphoric acid discussed earlier (8). Dr. Faucher is also managing contract research being undertaken by the Amicon Corporation in Cambridge, Massachusetts, in which asbestos may have a prominent role. In this instance the function of asbestos as a nucleating agent for the growth of fibrous crystalline forms in the tobermorite gel portion of hydrated cement is being investigated (9).

Recent work by Mr. C. W. Schersten under the supervision of Dr. E. G. Caflisch at Olefins Research evaluated soft hydrocarbon resin as binders for Coalinga asbestos (10). They report several compositions in the 3100 - 3900 psi flexural strength range. These materials have possible applications such as floor tile, vinyl floor tile backing and insulation or building board used in the construction of frame houses.

B. Other Carbide Asbestos Projects

Two projects under the supervision of Dr. Ralph Stickle at the Technical Center in South Charleston utilize asbestos which has a slightly longer fiber than that obtained in Coalinga asbestos. The first project directed by Mr. C. E. Metten, involves work concerning latex binding of asbestos felt used as a flooring cover. This effort is in cooperation with Congoleum Nairn and utilizes what is thought to be a Canadian asbestos source. The second project involves the incorporation of Canadian asbestos into a weather-proofing mastic formulation. This work, directed by Mr. W. F. Hill, is fairly well advanced; a mastic formulation has performed very well in preliminary trials at the Institute plant. The recommended formulation includes two grades of Canadian asbestos (11).

Asbestos Chemistry and Physical Properties

Asbestos as a chemical has probably presented as much mystery to modern researchers as it did to the ancients. About thirty minerals of fibrous crystalline structure are classified in this group, but only six are of commercial importance. These are, in order of importance, chrysotile, crocidolite, amosite, anthophyllite, tremolite and actinolite (12). However, the group has been subdivided into two general classes according to the method of formation. These are (a) serpentine asbestos which was formed by the serpentinization of adjacent host rocks by the conversion of anhydrous to hydrous magnesium silicate (13), and (b) amphibole asbestos which is thought to have been formed by a reversal of the hydration process. Water analysis of serpentine and amphibole asbestos shows an average content of 14 per cent for the former and 2 per cent for the latter (14). Of the six types mentioned previously only crysotile is a serpentine asbestos, while the other five are classified as amphibole types (12).

Actual composition of the various types of asbestos appears to be fairly well established and is shown in Table I (15).

TABLE I

CHEMICAL COMPOSITIONS OF VARIOUS TYPES OF ASBESTOS

	Chrysotile	Crocidolite	Amosite	Anthophyllite	Tremolite
SiO_2	37 - 44%	49 - 53%	49 - 53%	56 - 58%	51 - 62%
MgO	39 - 44%	0 - 3%	1 - 7%	28 - 34%	0 - 30%
FeO	0.0 - 6.0%	13 - 20%	34 - 44%	3 - 12%	1.5 - 5.0%
Fe_2O_3	0.1 - 5.0%	17 - 20%	---	---	---
Al_2O_3	0.2 - 1.5%	---	2 - 9%	0.5 - 1.5%	1.4 - 4.0%
H_2O	12.0 - 15.0%	2.4 - 4.5%	2 - 5%	1.0 - 6.0%	0 - 5.0%
CaO	Tr. - 5.0%	---	---	---	0 - 18%
Na_2O	---	4.0 - 8.5%	---	---	0 - 9%
$CaO + Na_2O$	---	---	0.5 - 2.5%	---	---

Actinolite is similar in composition to tremolite except that iron replaces some of the magnesium (16).

Crystalline structure, however, has presented a riddle. Chrysotile was first imagined as a chain structure with the general formula of $3MgO \cdot 2SiO_2 \cdot 2H_2O$ (17), but electron microscopy indicated that the individual fibrils exist in the form of tiny hollow cylinders. These cylindrical fibrils have diameters ranging from 200 Å to 400 Å (18). As a measure of fineness human hair may have 630 fibrils to the linear inch compared to 850,000 to 1,400,000 fibrils to the linear inch for chrysotile (19).

Moreover, X-ray studies of chrysotile, indicate that the cylinders were actually made up of layers in the sequence of 3O, 2Si, 20 OH, 3 Mg, 3 OH (20). This shows that the water in chrysotile exists as hydroxyl (OH) ions, so that the present formula is thought to be $Mg_3(OH)_4Si_2O_5$ (13). Also, the studies showed that the layers, which are 7.3 Å thick, consist of silicon-oxygen tetrahedra condensed onto magnesium hydroxide layers. Fibril walls consist of 12 to 20 of these layers joined by mechanical, but no chemical, linkage (21). However, these studies suggest hollow tubes with an inside diameter of 100 to 200 Å and a void fraction of 25 per cent; density studies on the other hand show chrysotile as having a void fraction of about 6 per cent with pore radii in the range of 13 to 16 Å (22). Dr. E. J. W. Whittaker of Terodo Ltd., England and Dr. Fred L. Pundsack of Johns-Manville have tried to resolve this discrepancy by postulating a "stuffed tube theory" in which amorphous or partially oriented material of the same composition as the outer walls fills the cylinders (21,22).

Another point of contention concerning asbestos appears to be the resistance to acids and alkali. Dr. E. J. W. Whittaker predicts high alkali resistance by postulating a layer structure for chrysotile in which magnesium hydroxide layers are on the outside and silicon-oxygen tetrahedra on the inside of the fibrils (21). He also shows amphibole asbestos fibers to have layers which have the silicon-oxygen tetrahedra on the outside. This results in high acid resistance. However, a recent report by the Nuclear Division, "Asbestos Data Book" states that chrysotile has a poor alkali resistance as well as poor acid resistance; this directly contradicts Dr. Whittington.

Physical studies of asbestos fibers reveal more interesting properties. Fibril diameters have been discussed and a comparison was made with human hair. Similar ratios (19) exist between asbestos and most fibers including wool, cotton, rayon, nylon and rock wool. Tensile studies reveal that chrysotile has a maximum tensile strength of 824,000 psi as compared with cotton having 125,000 psi or nylon having up to 117,000 psi (23). Crocidolite, which is replacing chrysotile in high strength applications, is the strongest asbestos fiber. Comparisons of chrysotile, crocidolite and steel tensiles show chrysotile having about 83 per cent and crocidolite having an amazing 150 per cent of steel tensile strength (24). From the small size of the fibrils, one might expect asbestos to have a very large surface area. This is indeed the case, although here too there is a difference of opinion. One source (24) lists the surface area of chrysotile as 13 to 22 sq. meter/gram while another shows it to have a specific surface of 1.06 to 2.3 sq. meters/gram (25). The former figure is probably the most accurate since the latter would place asbestos near the same values calculated for wool or rayon (24).

Water sorption by chrysotile fibers at 100 per cent R.H. is in the order of 2.5 per cent (26). In comparison, cotton absorbs 3 to 4 times and rayon 6 to 7 times this amount of moisture.

The Canadian Mining and Metallurgical Bulletin (April, 1951) shows a fairly complete list of physical and chemical properties of the six commercial types of asbestos. Some of these are shown in Table II for chrysotile and crocidolite (27).

TABLE II

SOME PHYSICAL AND CHEMICAL PROPERTIES OF CHRYSOTILE AND CROCIDOLITE

	Chrysotile	Crocidolite
Color	Green, gray, amber to white	Blue
Flexibility	High	Good
Resistance to heat destruction	Good, brittle at high temperature	Poor, fuses
Fusion Point, °F	2770	2180
Specific Heat, Btu/lb/°F	0.226	0.201
Electric Charge	Positive	Negative
Chrystal System	Monoclinic & Orthorhombic	Monoclinic
Hardness (Mohs')	2.5 - 4.0	4.0
pH	9.2 - 9.8	--
Specific Gravity	2.4 - 2.6	3.2 - 3.3
Refractive Index	1.50 - 1.55	1.7 Pleochroic

Production and Consumption

During the 19th Century large deposits of asbestos ore were discovered in Canada and South Africa; in 1962 Canada led in the production of asbestos with 40 per cent of the world total (3). Other producers, in the order of their importance, include the U.S.S.R., (36% est.), the Republic of South Africa (7.2%), Southern Rhodesia (4.6%), China (2.9% est.) Italy (2%), the U.S. (1.7%), and Swaziland (1.1%) (3). The variety, chrysotile, constitutes about 95% of the total world production (12). The United States consumes about 24 per cent of the world production of asbestos fiber (3). A large amount must therefore be imported. The 1962 Minerals Yearbook published by the U. S. Department of Interior gives the figures of U. S. consumption in 1962 as shown in Table III (3).

TABLE III

1962 PRODUCTION AND CONSUMPTION OF ASBESTOS IN THE US

	Short Tons	Value
U. S. Production	53,190	$ 4,677,000
U. S. Imports	676,027	64,150,000
Raw Fiber Exports	2,949	598,000
Asbestos Product Exports	---	14,274,000
Apparent Consumption	726,268	---
World Production	3,055,000	---

The bulk of U.S. imports are received from Canada with the Republic of South Africa being the next largest supplier. Amosite, a type of amphibole asbestos, is found only in the Republic of South Africa in commercial quantities. Southern Rhodesia produces a type of low iron content chrysotile which is essential to the production of electrical insulating material for shipboard electric cables (17).

Most of the domestic asbestos is produced at the Vermont Asbestos Mines Division of the Rubberoid Company near Hyde Park, Vermont. It is believed that these deposits constitute the southern extremity of the vast Canadian fields in the Quebec area (17). Other U. S. Mines are located in Arizona, California, Oregon, North Carolina, Virginia and Wyoming (5).

Prices (1962) for asbestos fiber at the mine in Canada and the U.S. range from a high of $1,650 per ton for the longest fiber spinning grades to a low of $40 per ton for the shortest fiber mill grades. Import prices vary from $163 to $221 per ton depending on the type of asbestos and the shipping distance (3). Long fiber chrysotile used in spinning must be at least 3/8 inch long and is mostly produced at the Canadian mines near Quebec (17). Spinning fibers constitute a small fraction of the total production and are quite expensive.

Asbestos-Cement Manufacture

A. Present Manufacturing Methods

In the past few years more than two hundred patents concerning asbestos-cement have been granted in industrialized countries (28); in addition, numerous articles have appeared in various journals. As one would expect from such activity, a wide diversification of formulations, additives, cure procedures, and product utilization have resulted. However, until recently asbestos-cement manufacture has followed a rather simple series of steps; the only advancement in technology seems to have been in the field of automation. These steps might be listed as follows:

1. Asbestos fiber "opening" or refining.
2. Particle suspension in a dilute aqueous slurry.
3. Web or felt formation on screens.
4. Felt compression by rolling.
5. Product formation (sheet, pipe, shingles, etc.).
6. Compression, drying and curing.

Most asbestos fiber in its crude or mill form consists of bundles of the tiny fibrils, discussed earlier, which must be opened to obtain the maximum surface area. In the case of asbestos-cement it has been found that maximum adsorption of cement gains produces greater strength in the finished product. Moreover, this high degree of adsorption and adhesion depends not only on the mineral composition of the cement but also upon the specific surface of the asbestos fibers (29). Perhaps the most common method of opening the bundles is simple mechanical refining or crushing by various types of beaters or edge mills. However, this treatment reduces the fiber length. To prevent this, other opening methods such as the flotation process of Raybestos-Manhattan, Inc. and the Vortrap process of the Naval Research Lab (30) have been developed.

The next four steps in asbestos-cement manufacture are completely automated in modern plants. After the asbestos is open, the fibers, hydraulic cement (usually Portland cement) and additives (if any) are suspended in a 90 - 99.5 per cent aqueous slurry which is thoroughly mixed and homogenized. From there the slurry passes onto the screen of a sheet forming machine, such as a Fourdinier machine, on which a web or felt is formed as the water drains away. This web is further rolled and pressed to remove excess water and to bring the sheet to the required thickness (31). The next step depends on the end product desired. If sheet is to be made, the web passes onto a sheet forming machine in which it is cut to the proper dimensions; if the product is pipe, the web is wound on a mandril; and if the item is shingles, the sheet is cut, veneered and embossed (for a weathered effect). At the Kurtz Plant in Germany the asbestos-cement sheets are interleaved with flat pieces of sheet iron as they come off the sheet machine. Then these stacks are dried under a pressure of 100 to 600 kg/cm and stored to allow the cement to fully hydrate (32). Items may also be steam cured in an autoclave at, for example, a steam pressure of 130 psi for 16 hours. If steam curing methods are employed, the product has better properties when some form of finely divided silica is included in the mix (33). Asbestos cement products usually have an asbestos content of 10 to

20 per cent although this may vary considerably. If silica is incorporated in the mix, a typical formulation (34) might contain 50 per cent Portland cement, 30 per cent silica, 20 per cent asbestos, and a water/cement ratio of 0.25 to 0.60 in the finished product to allow for maximum hydration of the cement.

B. Recent Manufacturing Developments

A recent patent (British; No. 922,427; April 3, 1963) granted to the Johns-Manville Corporation describing a method of forming asbestos-cement products by extrusion shows considerable promise as a means of forming more complex items, such as multi-channel conduit pipe (34). In this disclosure the inventors extrude plastic or putty-like mixtures in which a minimum quantity of water (25 per cent of cement wt.) is bound into the mix through the mechanism of a "hydro-modifier" such as POLYOX water-soluble resin [poly(ethylene oxide)] or "Methocel" (methyl cellulose). Heretofore, extrusion methods had failed because of a condition called "dewatering" which occurs to the mix as molding pressure is applied.

Another patent (31) approaches the problem of complex configurations from a different viewpoint. In this invention the asbestos-cement web is fabricated in the conventional manner, but afterwards is dried to less than 5 per cent (by weight) water. This prevents cement hydration. Synthetic or natural rubbers may be added to increase strength and flexibility of the dried sheet. The resulting sheet, possessing sufficient tensile strength for handling, may be formed or molded into complex shapes. Afterwards, it may be re-wetted and cured to form a tough, strong, hydrated asbestos-cement product.

Asbestos-Cement Modifications

In the Johns-Manville patent (34) described above, many formulations varying the type of cement, type of hydro-modifier and final density have been listed. For example, successful extrusion mixes have been made using other types of hydraulic cements such as supersulphated slag cement, calcium aluminate cement, natural cement, pozzolana cement, lime and similar materials. Densities of the cured products are varied not only by changing the water content but also through the use of inert fillers such as stone flour, ground shale, kaolin and perlite. The patent also lists twenty-seven water-soluble natural and synthetic polymers which were tested as hydro-modifiers. Thus, an insight to the wide scope of research on asbestos-cement products may be gained just by studying one patent.

However, other researchers have examined different properties of asbestos-cement products such as impact resistance, water penetration, hardness, and electrical properties in an effort to effect their improvement. In the U.S.S.R. scientists report an improvement of flexural, impact, and compressive strengths in asbestos-cement concrete containing poly(vinyl acetate) (33). A recent (1959) German patent (35) claims an improvement in heat insulation, sound insulation and flexibility of asbestos-cement sheets through the use of asbestos fibers impregnated with synthetic or natural rubbers. To fabricate an asbestos-cement sheet for use as electrical insulation, a Japanese inventor developed and patented a process employing epoxy resins and hardeners (36). Others in Japan have been studying the effect of various admixtures such as pulp, glass fibers, rock wool, natural and synthetic resins, slag powders

and fly ash on the impact resistance and hardness of asbestos-cement sheets (37). They report improved impact resistance through the incorporation of natural resins.

In the United States a non-inflammable and highly water-proof structural board was developed and patented by the Rubberoid Company using a sulfate cellulose fiber and asphalt in conjunction with asbestos and cement (38). Another type of building material, which the inventor calls a "facing slab", is made up of two layers (39): the backing of the slab is composed of Portland cement, asbestos, ground limestone, and a water-proofing compound, while the face layer is a mixture of granules (such as burnt shale or crushed monument stone), Portland cement, short asbestos fibers and pigment.

CONCLUSIONS Chemical and physical properties of asbestos show it to be a unique and useful mineral, most of which is consumed in the manufacture of asbestos-cement pipe and sheet products. In the literature surveyed, the most used modifiers of asbestos-cement were natural and synthetic resins and polymers, but no mention was found of asbestos-cement systems in which monomers were polymerized with heat.

Although some of the research by Union Carbide is directed toward a share of the asbestos-cement pipe and sheet market, no work, other than that which may result from this study, is concerned directly with asbestos-cement systems. The wide scope and variety of research in this field indicates that many problems still exist which merit further study.

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