

In the latter stage, these crystalline or porous areas will be found to have assumed a certain uniform pattern and position in relation to each other and to the immediate solids in contact therewith and suspended therein, providing merely a uniform dispersion and suspension of these solids through the dimensionality of the mold. And this dispersion and suspension remains static and permanent through the curing period, resulting in a porous product in which both the suspension agents and the solids have reacted and combined with each other and in their respective positions to become a single integral pattern with the loss of the specific and individual identity of the suspension agent so far as the latter is concerned.

Thus, if the finely divided reactive silica and dispersed or dissolved lime, as lime in water, are dispersed through a large volume of water, and the particles (or solution) remained in such a wide state of dispersion with and by the preformed reacted lumps of or platy crystalline compounds of crystals and the resulting therefrom to hydrous calcium silicate is effected either platy or lumpy crystals of hydrous calcium silicate will be induced or compelled to grow and combine with the preformed crystals and present a dispersed, entangled mass of crystals throughout the entire volume into which the reactive agents (and such points of infection) have been held suspended during reaction. Moreover, the so-regarded forming of platy crystals will be the basis of their growth, since a portion of the old lattice of one of the growing crystals and with the preformed crystalline crystals and thus all these crystals will become interwoven to form an interlaid, open lattice of permanent arrangement and integrated structure following the pattern set thereby by the original suspension of the preformed crystals and solids in the water.

Upon completion of the crystal growing phase and subsequent removal of the uncombined water, the improved mass will present innumerable voids and air spaces which will be found to be continuous and capillary in character and intercommunicating, the result of which will be a product of continuous, filamentary, microscopically fine crystal formation having intervening small attenuated capillary air spaces between and separating them and with said spaces greater in volume than the solids.

The essence of this invention resides in the formation of a liquid slurry of reactive solids, held in suspension in the liquid by a suspending agent of the similar chemical identity as the solids, whereas the water-solids ratio is greater than 1:1, reacting the slurry by heat and/or pressure to form an integrated product wherein the suspending agent will integrate with the reactive solids to become an integral part thereof but with retention of the respective relationships of the agent and solids but with the

Further, it should be appreciated that there is a very distinctive difference between an "agent" and a "component" in that the former, when applied, becomes an integral part of the end product without any chemical or physical reaction between it and the integrated mass as a whole, without leaving any foreign agent or converted product therein, embedded or otherwise, and differing from the integrated end product and that which has a noticeable effect upon the various or particular characteristics of the product, as illustrated in the foregoing.

Modification may be reversed to within the spirit and scope of the associated theory.

2. The method of making light apparent density products from stable suspensions of finely divided reactive solids, comprising forming an aqueous slurry having dispersed therein reacted hydrous calcium silicate crystals in discrete particulate form, and finely divided reactive solids in the form of lime and silica, said discrete particles being crystals of fish-like shape, of nearly colloidal dimensions and having the chemical composition $5\text{CaO} \cdot 3\text{SiO}_2 \cdot \text{H}_2\text{O}$, said crystals having surface areas of approximately 45 to 54 square meters per gram of weight, mixing said slurry to uniformly disperse both the discrete particles and the solids through the liquid to thereby form a stable suspension thereof, the liquid to solids ratio being in excess of 1:1 by volume and reacting the reactive solids of the said slurry while in suspension by subjecting same to a temperature and pressure in excess of respectively 175°C. and 150 p. s. i. abt.

3. The method of making light apparatus density products from stable suspensions of finely divided reactive solids, comprising forming an aqueous slurry having dispersed therein reacted hydroxide cation silicate critical to discrete particle form, and finely divided reactive solids

The contents set forth above are also true with re-
spect to any other material in integrated form.

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In the light of the apparent density range of products herein contemplated, it will be found necessary to add to a mixture of lime, silica and water an amount of synthetic materials in a solid dispersible particle form, sufficient to support the weight of the products components in a fine stream of water, prior to the liquid medium, from the time of completion of the mixing operation until sufficient reaction has occurred to thereafter retain the components in their originally dispersed and suspended morphology.

the desired specific strength in discrete particle form is obtained by use of different surface specific surface areas and shapes of solid particles. The particles and their distribution within the matrix is promoted free and uniform dispersion throughout a volume of liquid upon being mixed therewith in dilute proportions (e. g. not in excess of 1% by weight). They will acquire and maintain a random and spaced apart arrangement throughout the fluid mixture with the resultant effect of random dispersion of the lime-silica solids and will induce incipient crystal formation at and from such spaced apart points as well as from the dispersed solids under reaction temperature and pressure and form with the converted solids an integrated porous structure. These added previously reacted synthetic crystalline compounds, because of their scale or lath-like shape, nearly colloidal dimensions and high specific surface areas, will form an interlaced and interlocked haystack lattice system, capable of supporting the finely divided solids of the mixture in the liquid, and be used in this open three-dimensional network or lattice system the reactive materials, such as finely divided free lime and silica, or lime and silica in other forms, are capable of dispersion without destroying either the lattice system or the suspension provided by these pre-formed hydrous calcium silicate crystals.

Thus, in the production of a particular integrated product, such as, synthetic xenonlite in integrated form, there is provided an aqueous slurry composed of added unreacted lime and unreacted silica, added reacted synthetic hydrous calcium silicate crystals containing lime and silica in a molar ratio of 1:1 and added hot water (150-175° F.); the added lime and silica being mixed together with the added reacted synthetic crystals and dispersed and suspended in and upon the lattice system formed by the added reacted synthetic crystals and with a ratio of water to total solids greater than 1:1 and wherein the total CaO and SiO₂ of the added solids and that of the crystals is in a molar ratio of 1:0.

After complete mixing of the above subject slurry, it is poured into molds or otherwise shaped and subsequently subjected to reaction temperatures and/or pressures for a period of time sufficient to react all of the component materials, including the silicate crystals into an inorganic or combined form, and then dried.

In this particular instance, where the molar ratio of the total lime to total silica is approximately 1:0, the molded sherry is subjected to a predetermined sintering temperature and pressure in excess of 175° C. and 130 p. s. i. atm. in an autoclave for a period of time varying downwardly from approximately 15 hours depending upon the pressure being utilized.

In this knowledge system it is necessary, in order to produce a given end product, to start with a specific theory

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#1. Dioxine (oxydant puissant)

Moisture..... 11.
 Temperature..... approximately 20° C.
 Pressure..... 27.2 p. s. i. abs.
 Time..... 8 hours.

= 2. Least (non)volatile compound)

Molar ratio..... 1:1
Temperature..... approximately 175° C
Pressure..... 110 mm
Time..... 8 hours

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Molar ratio..... 65:100:1
Temperature..... 140° C
Pressure..... 1500 p. s. i.
Time..... 12 hours

As a further example of the utilization of this invention, a mixture of 100 parts by weight of a compound which may be represented by the formula $\text{C}_6\text{H}_5\text{C}_6\text{H}_4\text{C}_6\text{H}_5$ and 100 parts by weight of a substance which is crystalline compound itself containing lime and silica in a molar ratio of 6 to 1, to produce either a pure compound or a combination of the particular compound and another hydrous calcium silicate.

#1 21 p. 2-4 '5 mnc=3/1

	Pounds
Sulphuric acid (10%)	350
C.S. water (10%)	
Lime	1066
Silica	1760
Water	9500

22. 11 p. c. 1 — W'S ratio = 6/1

	Pounds
Subsistence Agent (Hospital)	255
C. Smith, F. L.	
Lime	358
Soda	98
Diatomaceous earth	378
Water	6700

If the suspension agent is the crystalline compound zeonlite having lime and silica in the molar ratio of 1:1 the following composition will produce a pure crystalline product having the molar ratio of lime to silica of 1:1 therein.

23.7 p. c. f.—W/S ratio=8/1

Suspension agent (zincite).....	1600
C/S ratio 1.0:1.0:	
Line	230
Silica	2500
Water	1400

Other formulations having various lime to silica ratios of at least 0.65:1 to about 1:1 may be utilized in various water to solids ratio to produce products having densities ranging from the lightest p. c. f. in integrated form up to the point where the water to solids ratio is such as to obviate the need of any suspension of the reactive solids. The quantity of the particular reacted crystalline form of suspension agent required will vary with the desired density of the end product and in the entire range of products will be in a range of from approximately 3% to approximately 30% of the total solids of the slurry by weight, i. e., the solids including the lime, silica and the suspension agent. Such products may be made as pure crystalline compounds, a combination of compounds, a combination of a compound or compounds and Shown crystal structure.

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METHOD OF MAKING INTEGRATED LIME-SILICA PRODUCTS

George L. Malachuk, Toledo, Ohio, assignor to Owens-Illinois Glass Company, a corporation of Ohio

No Drawing. Application April 3, 1952.
Serial No. 286,297

9 Claims (Cl. 104-120)

In the production of various types of products in the lime-silica-water system, many attempts have been made to produce integrated products of light apparent density and some measure of success has been attained but in many instances, either the particular process followed or the particular materials utilized, or both, limited the degree of success.

In the manufacture of such integrated products within this system, the art, both patent and industrial, has had recourse to various methods of retarding the settling of solids in their liquid carriers, such as the use of bentonite gels, cooling, use of starch, etc. None of these, however, provide a permanent suspension, i. e., they retard the rate of settling of the solids but never provide a slurry having a stable suspension of the solids in a liquid carrier wherein the characteristics thereof carry over into the final integrated product. It will be found in these products that density varies thru the thickness of the product thus presenting a product of nonuniform density.

The use of either bentonite or powdered aluminum permits the production of comparatively light density products but the use of these materials as retardation or levitating agents for the solids in the slurry contributes detrimental factors that result in a limitation of the ultimate use of the products.

A new theory of suspension, as distinguished from retardation, has been described in the recent Fraser Patent 2,469,379 wherein highly spiculated asbestos fibers provide a system of suspension for the solids in a liquid carrier, which suspension is permanent to the extent that the original dispersion, suspension and the suspension agent retain their respective suspended position in the final product. However, this agent, namely, finely microspiculated asbestos fiber is non-reactive and retains its identity thereby presenting, for some purposes, an unsightly product and a product incapable of accepting a smooth surface finish.

There are at least two distinct differences between the old retardation methods and that of the Fraser patent, supra, namely, in the old methods the agents of retardation are converted and are only present in the ultimate product in their converted form whereas in Fraser, the agent is not converted but retains its original identity and remains imbedded in the end product in its original position.

All of these above discussed methods of retardation and suspension present in the end product some detrimental characteristic, for example, bentonite when used in quantities sufficient to provide a degree of retardation capable of producing a light density product, produces a very much weakened product of nonuniform density, while the above mentioned suspension of Fraser, although capable of producing a product of extremely light and uniform density with high modulus of rupture, also produces a product wherein the exterior surfaces are fuzzy and incapable of receiving or accepting a completely smooth surface finish.

In the present invention, the primary object is to pro-

duce a product of extremely light uniform density, high modulus of rupture and having the capacity to accept a high surface finish.

A further object is the production of an integrated product in the light apparent density range in which suspending is a matter of the solids, namely, lime and silica is necessary and wherein the suspension agent is a crystalline compound that loses its specific or individual identity in the end product.

Other objects will be apparent from the following specification.

To obtain the product of the present invention, it is necessary to have as the agent of suspension a crystalline compound in discrete particle or crystal form, each crystal having specific surface areas capable of rendering a high degree of dispersion and stable suspension of the solids in a liquid medium.

A further distinctive feature in the production of this type product is that the agent of suspension becomes an integral part of the final integrated product and loses its specific individual identity.

Although the preferred feature of this present invention is the formation of a permanent suspension for making integrated products, it is not beyond the purview of the invention to use these crystalline compounds for obtaining suspensions of various other solids in liquid mediums of various types and in particular, where it is requisite that the agent of suspension should lose its specific apparent identity in the end product.

Further, it is the purpose of the present invention to provide not only a stable permanent suspension of the solids in a liquid carrier but to obtain such suspension with an agent the dimensions of which are of colloidal or nearly colloidal order and which will lose its specific and individual apparent identity in the finished product and permit the final product to accept a smooth surface finish.

Such an agent may be the product in discrete particle form obtained in accordance with the invention of the corresponding application Serial No. 12,452, filed March, 1948, now abandoned owned by a common assignee and which is directed to the manufacture of Lepisl, $4\text{CaO} \cdot 5\text{SiO}_2 \cdot 3\text{H}_2\text{O}$, in discrete particle form.

Lepisl in discrete particle form, is an extremely thin scale or plate-like shape having specific surface areas ranging approximately from 48 to 55 square meters per gram of weight.

Other similar agents, such as synthetic sonotile, $5\text{CaO} \cdot 5\text{SiO}_2 \cdot 3\text{H}_2\text{O}$ in discrete particle form, because of its lath-like form and also its high specific surface areas, approximately 45 to 54 square meters per gram of weight, may also be used as a suspension agent and there may be other silicate compounds or structures having the necessary attributes for this purpose but as the specific example in this present specification, sonotile shall be referred to as the agent for suspension.

Suspension of the finely divided lime and silica solids as contemplated in this invention is only necessary in the light density field, i. e., wherein the apparent density ranges from the lightest up to approximately 40 p. c. f. or wherein the ratio of liquids to total solids is greater than 1:1 by volume.

In order to produce a particular lime-silica integrated product, for example, sonotile ($5\text{CaO} \cdot 5\text{SiO}_2 \cdot 3\text{H}_2\text{O}$) in accordance with this invention, a specific molecular ratio of lime to silica must be present in the slurry, which in this instance, is 1:1.

These lath-like silicate crystals of synthetic sonotile, which provide this basis for suspension of the solids in a liquid, each contain lime and silica in a molar ratio of 1:0 and thus provide in the slurry a predetermined weight of these two reactive components. The solid lime and silica either in free or combined form must be added in

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which first made the use of a vacuum in which
the air was removed and the process was
GEORGE L. HALLIDAY.

REFERENCES CITED

Being references to the state of the art in the
art of the patent:

UNITED STATES PATENTS

Number	Name	Date
2,155,001	Smith et al.	Sept. 24, 1940
2,155,002	Smith et al.	June 2, 1940
2,155,003	Smith et al.	May 10, 1949

FOREIGN PATENTS

Number	Country	Date
2,155,001	Great Britain	Oct. 1, 1940
2,155,002	Great Britain	Oct. 1, 1940

OTHER REFERENCES

Dana's Textbook of Mineralogy, 4th ed., re-
vised, New York, New York, John Wiley and
Sons, 1932, page 641.

From this, it is evident that the Government has
this new method of dealing with the people.
During the past few years, the Government has been
trying to develop a new method of dealing with the
people.

The formation of this pure microcrystalline both structure is actually a transition of the important molecular stage - gel - and water, from the solid or vitrified first to a time for formation, then to an extremely fine crystalline structure but still retaining the properties of a gel, then to a continuation of this last mentioned second crystals of both-like structure in formed clusters, the crystallization of the whole to a pure microcrystalline both structure in formed clusters in random direction through the mass with approximate complete elimination of any gel formation, with the complete crystallization upon drying of all uncombined water and the formation thereby of voids in aggregate volume in excess of the aggregate volume of the both-like crystals, the both crystal beams of parallel extinction, positive elongation, having number of reflections slight exceeding 100, wavelength 1.071-1.072, the time after and water bound in the crystals in the molar proportions of 5, 6, 1, respectively, this transition for practical and commercial purposes being effected through the application of pressure, the temperature which will vary in the case of different materials, and the method employed, and means of crystallization used to complete the transition to a uniform and solid product.

The term "unit" as used herein shall be interpreted as meaning a mole ratio of 1:1 in the crystals of the fused product.

1. A solid solution of polymer and a solid body structure can be a function of time, temperature, size of crystals, and the formula.

and have been closely integrated with each other and with the local community. The two former are the mainstay of the national defence effort and the latter are the backbone of the national economy.

3. The method of forming an integrated product which comprises forming an intimate reactive mixture consisting essentially of lime, silica, and water, the amount of water in the mixture being within the range of about 4.5 to .75 times the approximate weight of the solids, the molar ratio of the reactive CaO and SiO₂ being approximately 1:1, shaping the mixture, subjecting the shaped mixture to a predetermined hydrating temperature and pressure in excess of 173° C. and 125 p. s. i., respectively, and thereby causing a reaction by which the lime, silica and water are converted completely to an integrated porous structure of pure microcrystalline lath crystals, having the formula 5CaO·4SiO₂·3H₂O, said crystals being in random dispersion and being integrated with each other and thereby maintaining said structure in integrated form independently of any other bonding medium, and drying the product to

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In addition to the above factors, it is found that in the field of compressive strength, this new product is much superior in its field, as 20 = density will have a compressive strength of 500 = 1000 psi, 30 = density 600 = 1000 psi, and 45 = density 800 = 1000 psi.

For example, this material at 212 degrees was subjected to a series of tests wherein the surface temperature, from direct flame application, reached approximately 2000 F. and after two hours of exposure thereto was subjected to a stream of cold water under pressure without chattering and with only superficial surface cracking.

The advantage of this material is extremely low when compared to other lime-silica products and its hardness of 4.5 according to Mohs scale. Together with its number of with-standing extreme thermal shock upon quenching makes it a most desirable product for many purposes, and in particular for fire resistant structures. Such a product, because of its rigid structure lends itself to various forms or methods of finishing, such as:

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The nature of the carriers to be used will be a function of the apparent density of the binder, ρ_b , and ρ_c . For the lower densities of the binder, about 1 to 1.5 g./c.c., a suspension and each one of the aforementioned carriers is recommended, and a mixture of carrier and water and silica in which the weight ratio of the CaO to that of the SiO_2 is approximately 1:1. The maximum is because complete wetting of the carrier if too hot water is added. The water stirring continuously and the binder mixture for 1 or 2 minutes. The amount of carrier is parts per part of binder, and binder from about 10 for the

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10 11. The method of making light apparent density products from stable suspensions of finely divided reactive solids, comprising forming an aqueous slurry having dispersed therein hydrous calcium silicate crystals in discrete particle form, and finely divided reactive solids, said discrete particles being reacted crystals of both a needle-like shape, of nearly colloidal dimensions and having a chemical composition wherein the molar ratio of lime to silica is 1:0 or less, said crystals having surface areas of approximately 45 to 55 square meters per gram of weight, and crystals being selected from the group consisting of crystals having a chemical composition of $4C_2O_3Si_2O_5 \cdot 2H_2O$ and $4C_2O_3Si_2O_5 \cdot 2H_2O$, and being present in a slurry in a molar ratio within the molar range of at least 0.65:1 to about 1:1, mixing said slurry to uniformly disperse both the discrete particles and the solids through the liquid to thereby form a stable suspension thereof, the liquid to solids ratio being in excess of 1:1 by volume, and reacting the reactive components of the said slurry while in suspension by subjecting same to a temperature and pressure in excess of respectively 175° C. and 130 p. s. i. abs.

15 12. The method of making light apparent density products from stable suspensions of finely divided reactive solids, comprising forming an aqueous slurry having dispersed therein hydrous calcium silicate crystals in discrete particle form, and finely divided reactive solids, said discrete particles being reacted crystals of both a needle-like shape, of nearly colloidal dimensions and having a chemical composition wherein the molar ratio of lime to silica is 1:0 or less, said crystals having surface areas of approximately 45 to 55 square meters per gram of weight, said crystals being selected from the group consisting of crystals having a chemical composition of $4C_2O_3Si_2O_5 \cdot 2H_2O$ and $4C_2O_3Si_2O_5 \cdot 2H_2O$, and being present in a slurry in a molar ratio within the molar range of at least 0.65:1 to about 1:1, mixing said slurry to uniformly disperse both the discrete particles and the solids through the liquid to thereby form a stable suspension thereof, the liquid to solids ratio being in excess of 1:1 by volume, and reacting the reactive components of the said slurry while in suspension by subjecting same to a temperature and pressure in excess of respectively 175° C. and 130 p. s. i. abs.

20 13. The product prepared in accordance with the method set forth in claim 1.
14. The method of making light apparent density products from stable suspensions of finely divided reactive solids, which method comprises forming an aqueous slurry having dispersed therein reacted hydrous calcium silicate crystals in discrete particle form, and finely divided reactive solids of lime and silica, said discrete particles being of both a needle-like shape and having colloidal dimensions and having a chemical composition wherein the molar ratio of lime to silica is 1:0 or less, said discrete particles having surface areas of approximately 45 to 55 square meters per gram of weight, the molar ratio of lime to silica being 0.8:1, mixing said slurry to uniformly disperse both the discrete particles and the solids through the liquid to thereby form a stable suspension thereof, the liquid to solids ratio being in excess of 1:1 by volume, and reacting the reactive solids of said slurry by subjecting same to a temperature and pressure in excess of 175° C. and 130 p. s. i. abs., respectively.

25 15. The product prepared in accordance with the method set forth in claim 14, said discrete particles of reacted hydrous calcium silicate being unidentifiable in said product.

16. The method of making light apparent density products from stable suspensions of finely divided reactive solids, which method comprises forming an aqueous slurry having dispersed therein reacted hydrous calcium silicate crystals in discrete particle form, and finely divided reactive solids of lime and silica, said discrete particles being of both a needle-like shape and having colloidal dimensions and having a chemical composition wherein the molar ratio of lime to silica is 1:0 or less, said discrete particles having surface areas of approximately 45 to 55 square meters per gram of weight, the molar ratio of lime to silica being 0.8:1, mixing said slurry to uniformly disperse both the discrete particles and the solids through the liquid to thereby form a stable suspension thereof, the liquid to solids ratio being in excess of 1:1 by volume, and reacting the reactive solids of said slurry by subjecting same to a temperature and pressure in excess of 175° C. and 130 p. s. i. abs., respectively.

17. The product prepared in accordance with the method set forth in claim 16, said discrete particles of reacted hydrous calcium silicate being unidentifiable in said product.

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18. The method of making light apparent density products from stable suspensions of finely divided reactive solids, which method comprises forming an aqueous slurry having dispersed therein reacted hydrous calcium silicate crystals in discrete particle form, and finely divided reactive solids of lime and silica, said discrete particles being of both a needle-like shape and having colloidal dimensions and having a chemical composition wherein the molar ratio of lime to silica is 1:0 or less, said discrete particles having surface areas of approximately 45 to 55 square meters per gram of weight, the molar ratio of lime to silica being 1:1, mixing said slurry to uniformly disperse both the discrete particles and the solids through the liquid to thereby form a stable suspension thereof, the liquid to solids ratio being in excess of 1:1 by volume, and reacting the reactive solids of said slurry by subjecting same to a temperature and pressure in excess of 175° C. and 130 p. s. i. abs., respectively.

19. The product prepared in accordance with the method set forth in claim 18, said discrete particles of reacted hydrous calcium silicate being unidentifiable in said product.

20 20. The method of making light apparent density products from stable suspensions of finely divided reactive solids, which method comprises forming an aqueous slurry having dispersed therein reacted hydrous calcium silicate crystals in discrete particle form, and finely divided reactive solids of lime and silica, said discrete particles being of both a needle-like shape and having colloidal dimensions and having a chemical composition wherein the molar ratio of lime to silica is 1:0 or less, said discrete particles having surface areas of approximately 45 to 55 square meters per gram of weight, the molar ratio of lime to silica being 0.8:1, mixing said slurry to uniformly disperse both the discrete particles and the solids through the liquid to thereby form a stable suspension thereof, the liquid to solids ratio being in excess of 1:1 by volume, and reacting the reactive solids of said slurry by subjecting same to a temperature and pressure in excess of 175° C. and 130 p. s. i. abs., respectively.

21 21. The product prepared in accordance with the method set forth in claim 20, said discrete particles of reacted hydrous calcium silicate being unidentifiable in said product.

References Cited in the file of this patent

UNITED STATES PATENTS		
Re 23,228	Fraser	May 5, 1950
2,315,891	Thomas et al	Sept. 24, 1940
2,340,774	Golden	Feb. 6, 1951
2,347,127	Kalousek	Apr. 3, 1951