106. COMPOSITIONS, COATING OR PLASTIC.

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CHESTER L. DAWES AND WILLIS A. BOUGHTON, OF CAMBRIDGE, MASSACHUSETTS, ASSIGNORS TO NEW ENGLAND MICA COMPANY, OF WALTHAM, MASSACHUSETTS, A CORPORATION OF MASSACHUSETTS.

## COMPOSITE INSULATING BODIES AND METHOD OF PRODUCING THEM.

No Drawing. Application filed October 26, 1922, Serial No. 597,154.

*To all whom it may concern:*

Be it known that we, CHESTER L. DAWES and WILLIS A. BOUGHTON, citizens of the United States of America, and residents of Cambridge, in the county of Middlesex and State of Massachusetts, have invented new and useful Improvements in Composite Insulating Bodies and Methods of Producing Them, of which the following is a specification.

Our invention relates to the manufacture of composite insulating plates or other bodies comprising mica flakes or fragments and a binding material which adhesively integrates the composite assemblage, and the object of the invention is to produce insulating bodies of this character by utilizing as binding material silicates of the alkali metals, either singly or in composition, and either with or without mixture with other salts. While in some aspects applicable to the production of silicate-bound mica-fragment insulators capable of service at only moderately high temperatures, our invention has for its main object the production of high-heat and mica-flake insulating bodies, capable of efficient service up to the disintegration temperature of mica such as we have had under observation (about 700°–750° C.) which sets a maximum temperature limit to insulators comprising mica of this variety.

Besides the mica-flake plates bound with an organic material such as shellac, which disintegrates at temperatures considerably below that which mica will endure, mica-flake insulators have been produced and used to some extent, in which the flake material is bound with a silicate, such as sodium silicate; but these have proved defective because of the large content of water remaining in the silicate, which at temperatures sufficiently high to produce steam causes the binding material to intumesce, seriously impairing the physical integrity of the insulator and also its insulating efficiency, because of local recondensation of steam.

In order to attain maximum efficiency in a built-up mica-flake plate, the adhesive binder used to integrate the mica fragments into a mechanically strong body must at ordinary pressures, be practically a solid at temperatures up to that at which mica disintegrates, because fluidity of the binder at lower temperatures will be accompanied by mobility of the mica fragments. On the other hand, in order to manufacture an adhesively integrated assemblage of mica fragments, the binder employed must be such as to flow at temperatures below the mica-disintegration point. Thus, in its principal aspect, the means of obtaining a reconciliation between the physical requirements for manufacture on the one hand, and for efficiency in use, on the other, constitutes or characterizes the invention herein described.

The liquefying temperatures of solid or anhydrous alkali-metal silicates, following a law of very wide application, become less with increase of pressure, usually provided the pressure is not hydrostatic, i. e. equal in all directions. This is true of these silicates whether liquefaction be true fusion, or due to reduction in viscosity of a silicate which retains a small residue of water and may be in a condition of colloidal suspension. Thus, by selecting and manipulating alkali-metal silicates either as single compounds or as mixtures, which under ordinary pressure and at temperatures near to the disintegration point for mica are practically solid, while they are liquefiable or can be made to flow at or about the same temperature provided increased pressure is applied to them, it is possible to construct a mica-flake insulator, integrated by a silicate binder which at ordinary pressures remains practically solid up to the temperature at which mica disintegrates.

Actual fusion of a silicate involves the expulsion of all water previously retained in association and requires a very elevated temperature. Since they can be made to flow, and thus to manifest all the characteristics of fusion necessary for the practical purposes of manufacture of mica-flake insulators, without expulsion of all water, at more moderate temperatures or pressures than those demanded for true fusion, the alkali metal silicates may in practice be advantageously used in this condition albeit that the water content is reduced to a relatively small percentage.

The simplest constitution of such silicates (represented by $Na_2SiO_3$) is seldom real-

ized; the $Na_2O$ and $SiO_2$ factors vary widely, and we have found that the silicates of the alkali metals in which the silica factor is high (as for instance a 3 to 1 or 4 to 1 ratio between $SiO_2$ and $Na_2O$) serve better for purposes of binding mica-flakes than those in which the ratio is lower in $SiO_2$. If mica flakes of the variety we have employed in our research and a high-silica alkali-metal silicate be assembled in a suitable press (the silicate either in a flowing solution or practically solid at ordinary temperature and pressure, although containing a variable amount of water, and pulverized) and be raised to a temperature, say of 650° to 700° C., and there maintained until only so much water remains in association as is consistent with the temperature condition, and if pressure be applied to the assembled materials while maintaining the said temperature, and be increased, a pressure-factor which depends on the temperature actually maintained will presently be developed which represents the flowing pressure for the given temperature and the silicate will liquefy and flow, spreading between and over the mica flakes while the whole mass of material becomes condensed by expression of surplus binding silicate if the original quantity was in excess of that required to cover the mica flakes with thin adhesive films. On release of pressure, or subsidence of temperature, or both, the binder solidifies, and a mica-flake plate results which will thereafter retain its integrity in use under any temperatures short of that at which mica itself disintegrates.

Mica-flake plates, comprising silicate binders which, while analogous in composition to those high-silica compounds which require the application of pressure in order to flow at temperatures below the disintegration point of mica, are nevertheless relatively low in silica (by ratio) and capable of flowing at atmospheric pressure at similarly safe temperatures, will be found practically useful in many situations although incapable of retaining practically solid integrity at temperatures approaching that of mica disintegration. In treating the materials for mica-plate manufacture, comprising silicate binders of all the varying characteristics mentioned, these will advantageously be subjected to heat for a period sufficiently long to allow all except a small residue of water-content to escape, and then be subjected to pressure which, in all cases, serves to give its desired ultimate form and proportions to the mica-flake plate and to distribute the binding material over the surfaces of the flakes themselves, and which, in those instances which require pressure to produce a state of flux, serves to render fluid the binding material which otherwise would remain in a nearly, or practically, solid condition. The temperature applied should, in all cases, be substantially higher than that at which water boils; the generation of steam in the silicate binder often causes it to puff up; but if the steam representing the water is gradually liberated, there will be left only a small residue of water, which is not only harmless under conditions of subsequent use of the mica-flake plate, but also functions usefully in promoting the flow of the binder which is colloidally or chemically associated with it.

Alkali-metal silicates may be employed either as solutions, or in practically solid and comminuted condition, in the preliminary stages of manufacture of mica-flake plates, or they may be formed in the assemblage of materials by interspersing between the flakes two or more substances which under the conditions of heat, or heat and pressure, react to form the silicate which is ultimately to constitute the binder. For instance, a suspension of silica ($SiO_2$) in a solution of alkali hydroxide or carbonate, used as material for preliminary cementation of mica-flakes, dried and heated under pressure will produce a silicate binder. The degree of pressure required to effect flow of the binder-silicate at a safe temperature will depend largely on the silica ratio in the mixture and reaction. A typical illustration of the reaction is:

$$2NaOH+SiO_2=Na_2SiO_3+H_2O.$$

The synthetic production of alkali-silicate suitable for a mica-binder may be effected by heating and compressing admixed alkali salts of suitable nature with silica, by processes very similar to those used in commerce for manufacturing the alkali silicates.

*Composite alkali metal silicates.*—Mica-flake plates of excellent qualities, such as high electrical resistance, high dielectric strength, non-hydroscopicity mechanical strength when cold, resistance to atmospheric conditions, comparative freedom from corrosion or corrosiveness, and relative chemical stability under repeated temperature-fluctuations, are producible by the employment of composite silicates. A mixture of sodium and potassium silicates, employed as a binder and manipulated in the manner above described with respect to simple silicates, yields these results. Such a mixture may with advantage be produced synthetically by mixing pure potassium hydroxide, or carbonate, with a sodium silicate solution; the metathetical reaction which takes place when the mixture is treated by heat, or heat with pressure, is typically represented by:

$$Na_2SiO_3+2KOH=2NaOH+K_2SiO_3,$$

it being understood that the original quantity of sodium silicate is in excess, so that there remains after reaction a desired ratio

of sodium silicate and potassium silicate, with free caustic soda. The latter, however, will of course react with the excess silica in the original silicate to produce an alkali silicate of somewhat lower silica-alkali ratio.

Relatively small quantities of salts of the alkali metals or alkaline-earth metals, such as halogen salts, nitrates, sulfates, carbonates, oxides, arsenates, etc., which are comparatively infusible or non-liquefiable when alone, may be mixed with any of the binding materials, represented by the liquefiable salts of the alkali or alkaline-earth bases (which are characterized by liquefiability or reduced viscosity under various conditions of temperature and pressure), these fundamental binders being used either singly or in combination. Thus, alkali metal silicates, borates, or phosphates singly or in combination, mixed with the relatively higher melting other salts of the alkali or alkaline earth metals, yield composite binders of practically any desired liquefiability (with a view to temperatures and pressures employed) these mixtures with relatively higher melting substances have excellent properties of electrical resistance, and are thus adapted to the requirements of insulating plates or other shapes or bodies, and since, also, they react with, or become dissolved or suspended in, the predominant binder material when the application of heat and pressure produces liquefaction.

In any case, whether of a simple or composite binder, the manipulation will depend upon the condition in which the material is applied. If in solution or suspension, preliminary drying of the composite assemblage of mica flakes and binding material will in most cases be resorted to.

This is ordinarily done at relatively low temperatures and with pressures only sufficient to cause the flakes to adhere to one another and thus form a plate that holds together and can be handled. After this preliminary drying, the plate is subjected to high pressure and temperature. The maximum limit of temperature is imposed by the mica itself, which must not be exposed to a disintegration temperature; the pressure required to produce the requisite liquefaction of the binding material will vary according to the liquefaction temperature of the binder at ordinary atmospheric pressure. Tentatively, and without more confidence in their universal applicability than seems justified by the observations and data so far made and accumulated by our researches, we suggest the following as guides to the selection and manipulation of mica flake binding materials.

If a binding material, at atmospheric pressure, melts or liquefies at a temperature not higher than 800° C., it may be used alone, and be made to flow under pressures not difficult to apply in manufacturing practice. With unusually powerful pressure apparatus, which may possibly be designed, this suggested practical limit of refractoriness to heat may be raised.

If a binder-substance requires, at atmospheric pressure, a temperature in excess of 750° C., where mica such as we have had under observation disintegrates, pressure must be applied to make the binding material flow at safe temperatures below 750° C.

If substances are used, as ingredients in a composite binding material, which themselves are comparatively or actually non-liquefiable or infusible, they should be mixed with some inorganic salt with which (in solution or liquefaction analogous to fusion) they will react or dissolve to produce a resultant material which will flow under pressure, at temperatures consistent with the presence of mica. Water-insoluble salts may be used either dry and comminuted, or in suspension in water, or in a solution of or suspension in a soluble binder.

Bearing in mind that with some substances the temperature of liquefaction or flow becomes lower with increase of pressure, while with other substances, infusible at ordinary pressures at a given temperature, liquefaction or flow can be produced when sufficient pressure is applied, it may safely be stated; that to make a thoroughly satisfactory high-temperature mica-flake plate one should select an inorganic binder which is effectively adhesive, or becomes so at an available temperature (which is safe for mica) which, at ordinary pressures and at the temperature at which the plate is to be used, does not flow, at all events not enough to allow the plate to deform, decompose, or break up, under such use; that one should apply that binder in a distributed condition to the mica flakes; that one should raise the temperature as closely to the decomposition point of mica as is safe, to effect intended and desired chemical or physical changes or both; that one should then, or earlier, apply sufficient pressure to make the binder flow, fill the interstices, and adhere closely to the mica surfaces; that one should thereupon, while maintaining the applied pressure, lower the temperature to a point where, in spite of the pressure, the binder is in practically a solid state; that one should then relax the pressure and allow the plate to cool.

Examples, which we have found to yield practicable binders for high heat mica-flake plates, representing several of the groups of composite binders hereinabove discussed, are as follows:

Sodium silicate solution (highly viscous), 72 cc.; dry sodium carbonate, 2 g.; dry potassium carbonate, 2 g.; fused potassium hydroxide, 1.5 g.; water, 50 cc.

Sodium silicate, 72 cc.; potassium carbonate, 2.2 g.; potassium nitrate, 2 g.; magnesium oxide, 4 g.; water, 50 cc.

In the foregoing specification, mica-flake plates have been taken as an example of the insulating bodies, the production of which is the object of our invention; mica-flakes, however, should for the purposes of specification be taken as an example only, since mica in such comminuted condition as to be regarded as powdered will respond to treatment with binding materials of the character described. Insulating bodies having shapes or proportions other than those of relatively thin plates are, like true plates, capable of being manufactured by association of mica fragments and inorganic binders.

What we claim, and desire to secure by Letters Patent is:

1. An insulating body, mainly consisting of mica fragments and a silicate of an alkali metal containing substantially no water, compressively integrated.

2. An insulating body, mainly consisting of mica fragments and sodium silicate containing substantially no water, compressively integrated.

3. An insulating body, mainly consisting of mica fragments and a silicate of an alkali metal high in silica and containing substantially no water, compressively integrated.

4. An insulating body, mainly consisting of mica fragments and sodium silicate high in silica and containing substantially no water, compressively integrated.

Signed by us at Boston, Massachusetts, this nineteenth day of October, 1922.

CHESTER L. DAWES.
WILLIS A. BOUGHTON.