Oct. 2, 1962 G. SLAYTER 3,056,653

CONTROL OF CRYSTAL GROWTH IN MICA MATERIALS

Filed Nov. 18, 1959 4 Sheets-Sheet 1

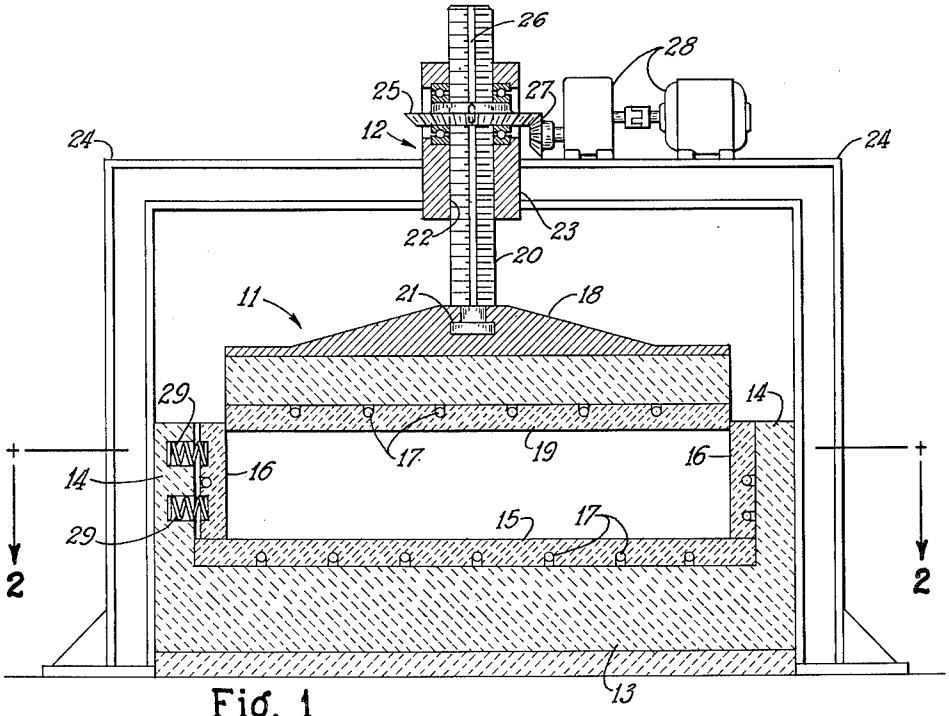


Fig. 1

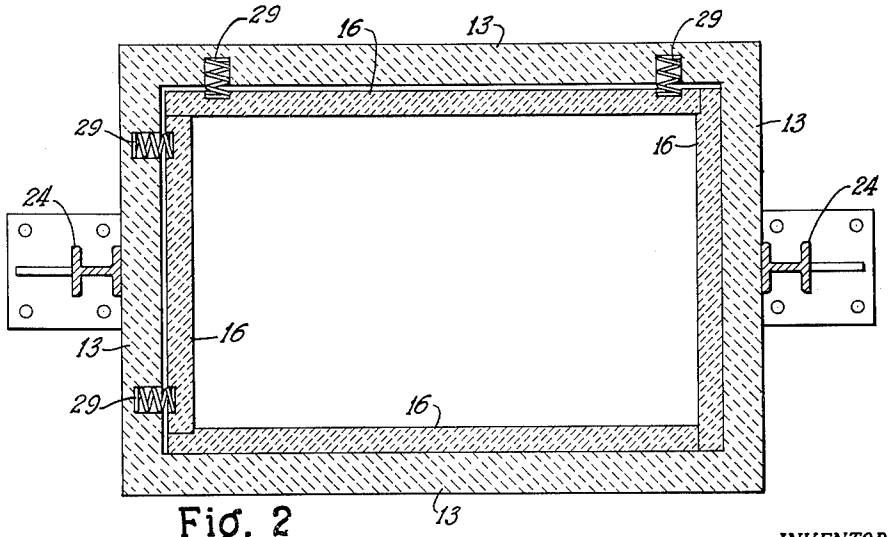


Fig. 2

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CONTROL OF CRYSTAL GROWTH IN MICA MATERIALS

Filed Nov. 18, 1959 4 Sheets-Sheet 2

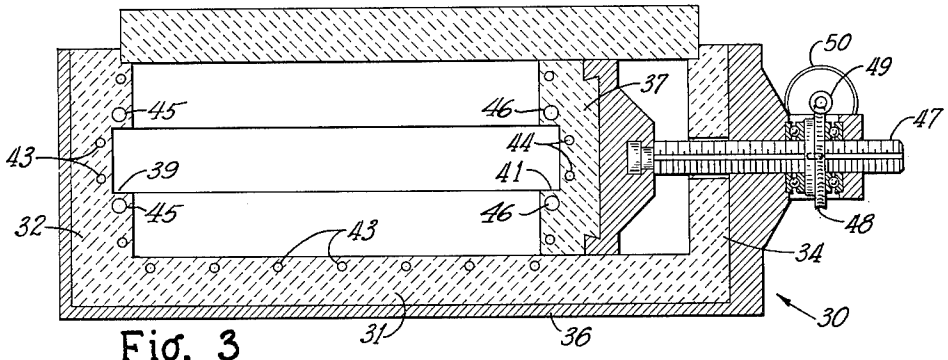


Fig. 3

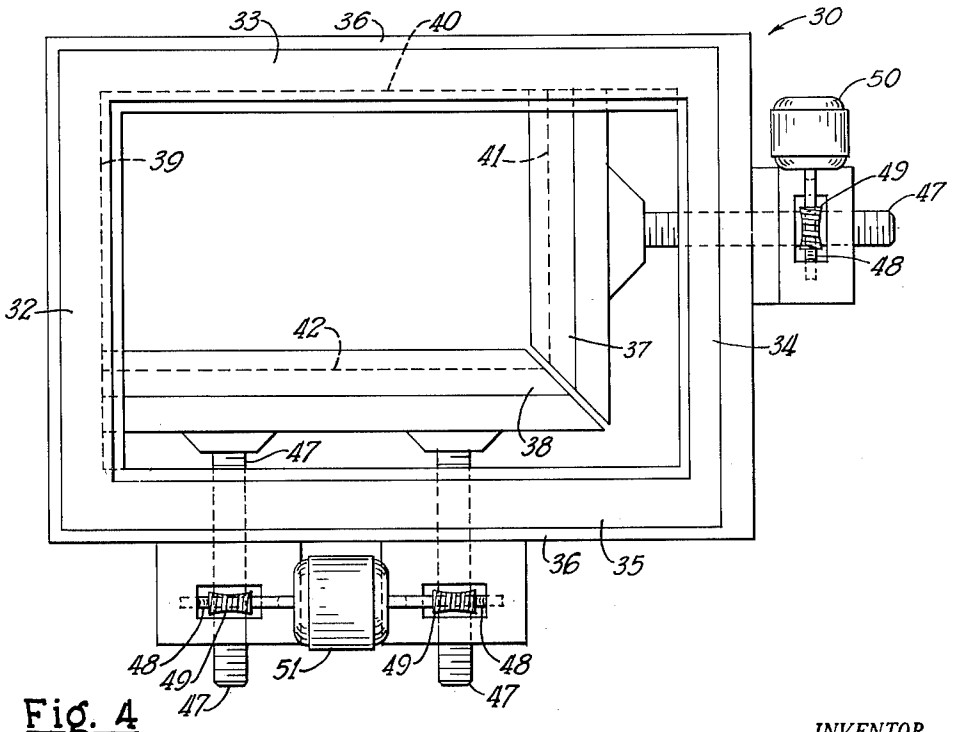


Fig. 4

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CONTROL OF CRYSTAL GROWTH IN MICA MATERIALS

Filed Nov. 18, 1959 4 Sheets-Sheet 3

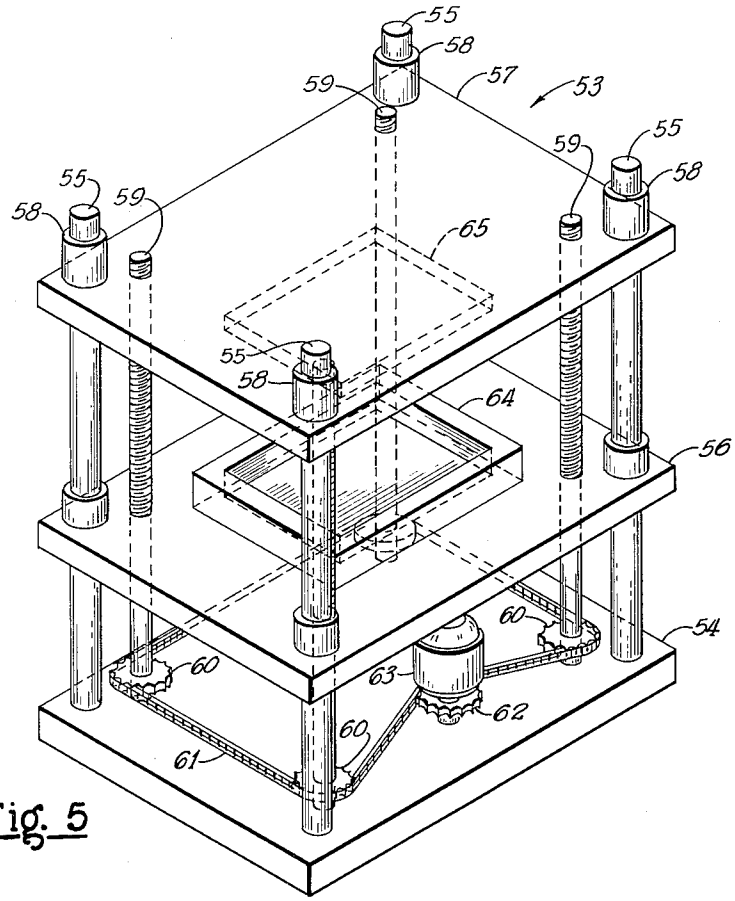


Fig. 5

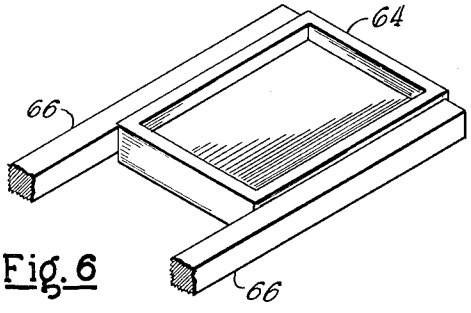


Fig. 6

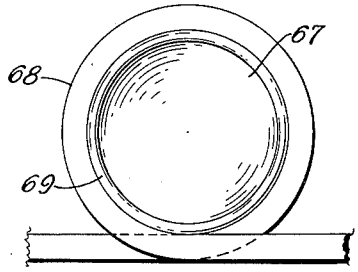


Fig. 7

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CONTROL OF CRYSTAL GROWTH IN MICA MATERIALS

Filed Nov. 18, 1959 4 Sheets-Sheet 4

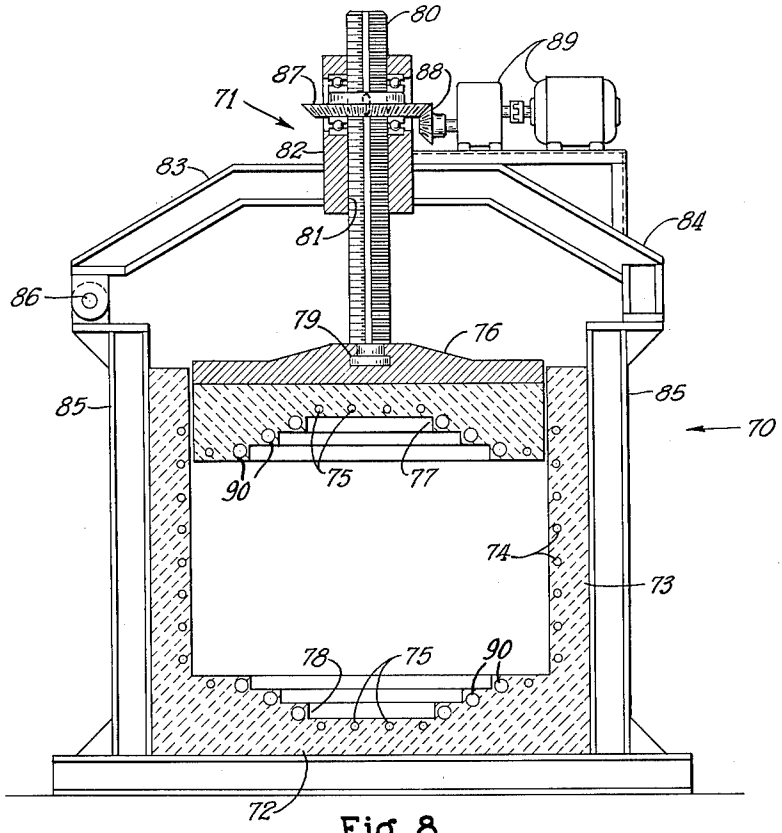


Fig. 8

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United States Patent Office

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3,056,653

CONTROL OF CRYSTAL GROWTH IN MICA MATERIALS

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Filed Nov. 18, 1959, Ser. No. 853,780
7 Claims. (Cl. 23—110)

This invention relates to the control of crystal formation in inorganic materials such as synthetic mica and more particularly to the formation of synthetic mica crystals by the application of controlled forces or pressure to molten synthetic mica as it cools.

Although mica formerly had rather limited uses it is gradually becoming more important. One reason for this increase in importance is the increased use of mica in aircraft, primarily brought about by the higher speeds attained by aircraft. Such speeds result in a greater amount of friction between the air and the skin of the aircraft with tempertaures above 1500° F. being frequently encountered. Although materials are now known which can effectively withstand these temperatures and which have been applied to the fuselages and wings of the high speed aircraft, no satisfactory material other than mica has been found for the canopies of such aircraft. Mica is ideal for this purpose because it is transparent and will withstand the high temperatures to which such canopies are subjected.

Most natural mica now used in this country is imported from India. Some domestic mica is available, but the cost of sorting it is excessive. Both domestic and foreign sources have been less than satisfactory, however, which has resulted in a search for improved methods of making synthetic mica. While having many other uses, synthetic mica is particularly adapted for high temperature applications because the physical properties of this mica can be controlled according to the composition employed and synthetic mica thereby can be made to withstand higher temperatures than natural mica.

Present synthetic mica is made by melting a suitable batch material and subsequently cooling it very slowly through a crystallization temperature range in which crystalline flakes or plates of the material are formed. The batch material can be melted in a crucible or piled on a furnace hearth with electrodes extending partially through the material, in which case the central portion of the batch pile melts and leaves an insulating crust thereover. Upon proper slow cooling, mica flakes are formed in the center of the molten portion as it solidifies. Such flakes are very small, being from ¼ to 3 inches long and constitute only a small portion of the total batch. The present synthetic mica is thus expensive, because only a small satisfactory quantity can be obtained in comparison to the size of the synthetic batch that is initially processed. Thus, the cost of labor, material, fuel, etc., has to be absorbed by a relatively small amount of the final product.

The present invention provides an improved method for making synthetic mica by means of which a greater quantity of larger crystals are obtained. This is accomplished by applying controlled force or pressure to the mica to place it in compression or tension while cooling. The pressure, depending on how applied, can actually produce a multiaxial, biaxial, or uniaxial stresses in the mica.

This causes the crystals formed to be larger than those heretofore produced, and to constitute a larger percentage of the total batch. Biaxial or multiaxial stress must be established in a common plane to produce larger mica flakes while uniaxial stress will be lineal to produce longer crystalline flakes and possibly even acicular or needle-like crystals in some instances.

Regardless of the direction, the pressure must be applied slowly and the synthetic mica batch must be cooled slowly. As it cools, some constituents tend to solidify or to crystallize more rapidly than others, a phenomenon known as fractional crystallization, which is common to a number of inorganic, glassy materials. As each constituent crystallizes, it does so under the influence of the pressure applied to the overall molten material and forms crystalline shapes dependent upon the nature of the stresses established by the pressure. The liquid constituents in the mica will continue to form in the particular shape depending on the stresses set up therein and the pressure, until all constituents solidify and there is no liquid left between the crystals to provide plastic flow by means of which the crystallizing constituents can travel in the mica under the influence of the pressure.

It is, therefore, a principal object of the invention to provide a method of solidification which will influence the formation of crystals in an inorganic, glassy substance.

Another object of the invention is to provide an improved method for producing synthetic mica flakes or sheets.

A further object of the invention is to provide a method of making larger crystalline flakes or sheets of synthetic mica.

Still another object of the invention is to provide a method of making synthetic mica in which a larger proportion of the mica batch is formed into crystalline flakes or sheets.

Other objects of the invention will be apparent from the following detailed description of preferred embodiments thereof, reference being made to the accompanying drawing, in which:

FIG. 1 is a view in vertical cross section of a furnace and force- or pressure-applying means for producing mica in accordance with the principles of the invention;

FIG. 2 is a view in horizontal cross section taken along the line 2—2 of FIG. 1;

FIG. 3 is a view in vertical cross section of another furnace employing modified means by which biaxial tensile forces can be applied to a molten mica batch, as it cools;

FIG. 4 is a top view of the apparatus shown in FIG. 3, with a cover removed;

FIG. 5 is a view in perspective showing apparatus for applying pressure to synthetic mica which has been melted in a separate furnace;

FIG. 6 is a detailed view of a pressure-applying block which can be employed with the apparatus shown in FIG. 5, with electrodes for heating the block;

FIG. 7 is a view of another pressure-applying block with modified heating means; and

FIG. 8 is a view in cross section of a modified furnace and pressure-applying means for placing mica under uniaxial stresses.

In the basic process of producing mica according to the invention, a suitable mica batch can be melted in a

furnace and pressure applied to the molten batch as it cools within the furnace. However, the molten batch also can be poured from the furnace into a recess or mold associated with pressure-applying apparatus. Pressure is then applied to the molten mica by the latter apparatus as the batch cools. In a second modification, mica from a furnace can be poured into a recess or mold and cooled. The cooled body can be subsequently placed in association with heated pressure-applying apparatus. The mica is then heated above its softening point and cooled slowly therethrough with pressure applied by this apparatus to establish uniaxial or biaxial stresses.

The pressure or force applied to the molten mica sets up uniform stresses therein to control the direction of movement of those portions of the mica which are still in a sufficiently fluid state to be mobile. When a compressive force or positive pressure is applied to place the mica under compression, crystal growth is generally perpendicular to the applied force whereas when tensile force or negative pressure is applied to place the mica under tension, crystals grow in a direction parallel to the tensile force. In the former instance with mica under compression, the crystals grow in a plane so as to form relatively large flakes or sheets. Because further stress is set up as portions of the mica solidify and change phase, the final resulting crystalline flakes or sheets are disposed in various planes according to the resultant of the composite stresses set up by the pressure and by the cooling. Thus, all of the crystals will not lie in the same plane. In the latter instance with the mica under negative pressure or tension, stresses set up therein are uniaxial and the crystals form in longer flakes or even in needle-like configurations. Crystalline flakes or sheets have the greatest application for such uses as aircraft canopies or wherever strong transparent material is desired, particularly where subjected to high temperatures. However, mica with needle-like crystals also has many uses particularly where reasonable strength at high temperatures is required.

In a specific form of the invention, by way of example, a batch is prepared consisting of

$$\tfrac{2}{3}K_2SiF_6+\tfrac{1}{3}K_2CO_3+6MgO+Al_2O_3+5\tfrac{1}{3}SiO_2$$

which yields the fluor-phlogopite formula

$$K_2Mg_6Al_2Si_6O_{20}F_4$$

A slight excess of fluorine is also used. This batch is heated to 2640° F. and soaked at that temperature for approximately 6 hours, depending on the quantity employed. It is then cooled in the furnace to a temperature of approximately 2515° F. which is the upper temperature of the crystal-forming range for this particular batch. Further cooling is then effected to approximately 415° F. at a slow, accurately controlled rate of 3.5° F. per hour. During at least the first one-third of this latter cooling period, a uniform positive pressure of 150 pounds per square inch is maintained on the batch.

FIG. 1 shows specific apparatus for carrying out the invention comprising a furnace **11** and pressure-applying apparatus **12**. The furnace **11** includes an insulating refractory floor **13** and side walls **14**, a carbon liner **15** being disposed on the floor **13** and a carbon liner **16** on the side walls **14**. Molten mica tends to stick less to these liners and contamination of the mica is also minimized. For some other synthetic mica batches, other liners may be preferred. These include silicon carbide, platinum, graphite (a form of carbon), and high silica fire clay.

The furnace **11** can be heated by a variety of means but preferably by some form of electrical heating to obtain closer control of the rates of heating and cooling of the mica, which rates are critical. In the embodiment shown, electrically insulated resistance elements **17** are imbedded in the carbon liners **15** and **16** or can be just below these liners in the refractory floor **13** and the refractory side walls **14**.

The pressure-applying apparatus **12** includes a pressure plate **18** and a suitable liner **19**, preferably of a carbon-containing material, which plate and liner serve as a cover while batch is heated and as a platen to apply pressure to the batch while it is cooling. The plate **18** is connected to a threaded shank **20** through a suitable rotatable joint **21** which enables the shank to rotate while the plate **18** remains stationary. The shank **20** extends through a threaded hole **22** in a block **23** which is affixed to suitable supports **24**, the shank being rotated by a bevel gear **25** slidably but non-rotatably held through a key **26**. The gear **25** meshes with a pinion bevel gear **27** driven by a motor and reducer combination **28**.

In operation, batch is supplied to the furnace **11** to approximately the height of the side walls **14**. The plate **18** and the layer **19** are then lowered into contact with the batch to form a cover over it during heating. The resistance elements **17**, including those located in the layer **19** or just behind it, are then supplied with current to heat the batch uniformly. After melting for a predetermined period of time, the batch is allowed to cool either by cutting off all current to the resistance elements **17** and allowing the batch to cool slowly in the furnace or by reducing the current to the elements **17** to reduce heat transfer to the batch and to enable it to cool even more slowly. As the batch cools, but while still in a molten or at least soft state, the motor **28** is operated to turn the shank **20** and to move the plate **18** and the liner **19** downwardly so as to apply a uniform pressure over the upper surface of the batch. Springs **29** are associated with two adjacent side walls **16** to enable the mica to yield outwardly while pressure is applied, particularly when the mica is relatively fluid. A constant pressure can be applied to the batch throughout at least an upper portion of the cooling range, pressure can be applied at a uniformly increasing rate, or pressure can be applied at an accelerated rate. One of the latter two may be preferred to enable more pressure to be applied to the batch as it cools further and becomes less fluid.

FIGS. 3 and 4 show a modified furnace **30** for establishing biaxial tension in mica by applying a negative pressure in two directions as the mica cools. The furnace **30** includes a refractory floor **31**, immovable side walls **32** and **33**, and retaining walls **34** and **35** in a metal casing **36**. Movable walls **37** and **38** constitute the other two walls of a space in which mica batch is placed. Recesses **39** and **40** are formed in the fixed walls **32** and **33** and recesses **41** and **42** are formed in the movable walls **37** and **38**, all of which recesses extend the lengths of their respective walls for reasons subsequently appearing. The side walls and bottom can have a liner of a suitable material compatible with the mica or can be made entirely of such material. Resistance elements **43** are disposed in each of the fixed walls **32** and **33** and the floor **31**, and resistance elements **44** are disposed in the movable walls **37** and **38**, the latter elements being supplied current through flexible leads. Adjacent the longitudinal recesses **39** and **40** are cooling tubes **45** through which a suitable coolant can be supplied and adjacent the recesses **41** and **42** are cooling tubes **46**, supplied with coolant through flexible conduits.

The movable walls **37** and **38** are driven in and out at right angles to each other by shanks **47** suitably threaded through reinforced portions of the retaining walls **34** and **35**. The shanks **47** have worm wheels **48** driven through worms **49** and a motor **50**, in the case of the wall **37**, and a motor **51** for the wall **38**, the latter motor driving the worm wheels **48** for both of the shanks **47** of the wall **38**. A cover **52** is also provided for the furnace.

In operation, the mica batch is placed in the furnace **30** and the cover **52** placed thereover. Heat is then applied to melt the batch and hold it in a molten state for the desired period of time. The power is then shut off or cut back and coolant is supplied through the tubes **45** to cool and to solidify the batch more quickly ad-

jacent the recesses 39–42. This portion of the batch thereby adheres more readily to the walls 32, 33 and 37, 38 to enable the batch to be placed under biaxial tensile stress as the walls 37 and 38 are moved away from the walls 32 and 33. The recesses 39–42 can be undercut somewhat to establish a greater holding power, if desirable. The biaxial tension or negative pressure applied to the mica in this manner establishes biaxial stresses which enable larger crystalline sheets of the mica to form as it cools. The walls 37 and 38 slide on the floor 31 with a close fit to prevent the possibility of any substantial leakage of the molten mica batch. Although the batch usually will be sufficiently solidified at the edges adjacent the walls 37 and 38 to prevent any substantial leakage between the junction of these walls, a flexible seal can be used at the junction, if desired. However, only a slight gap is actually formed thereat because the walls are not retracted extensively because only a relatively small movement is required to establish considerable biaxial stress. The walls 37 and 38 can be retracted to establish a constant pressure or stress, a constantly increasing stress, or a stress which increases at an increasing rate, as with the pressure-applying apparatus 12 shown in FIG. 1.

Rather than applying controlled pressure to the batch in the furnace, the molten batch can be poured from the furnace and pressure applied as the batch cools, or the batch can be cooled, subsequently reheated ,and the pressure then applied. FIG. 5 shows a press 53 by means of which mica can be subsequently placed under stress during cooling or after cooling and reheating. The press 53 includes a base plate 54 and four supporting and guide posts 55 at the corners of the plate. A stationary lower platen 56 is affixed to the supporting posts 55 and a movable upper platen 57 is slidably connected to the supporting post 55 by means of bearings 58. Four operating screws 59 are rotatably supported by the base plate 54 and extend vertically through the lower platen 56 and are engaged with threaded holes in the upper platen 57. The four screws 59 are turned in unison by gears 60 which are connected by a chain 61 driven by a drive gear 62 which is powered by a suitable motor and reducer combination 63. Blocks 64 and 65 which can be of carbon or other suitable material are located in alignment on the upper surface of lower platen 56 and on the lower surface of the upper platen 57, respectively.

As shown in FIG. 6, the block 64 can be heated by resistance through electrodes 66 which are connected electrically to opposite edges of the block. As shown in FIG. 7, a round block 67 can be heated by means of an induction coil 68 disposed around its periphery. The heated blocks enable the mica to be remelted after prior cooling or can be used to control the cooling rate of molten mica which is supplied directly to the blocks. Whether the blocks 64 and 67 are heated or not, they are preferably provided with lips 69 at their peripheries to hold the molten mica.

In operation, the mica is poured or placed on the lower block 64 or 67 which can be maintained at an elevated temperature along with the upper block 65. The gears 60 are then driven to rotate the screws 59 and lower the upper platen 58 and the upper block 65 to apply pressure to the mica, which is then allowed to cool with current to the plates 64 and 65 shut off entirely or cut back to retard the cooling rate.

FIG. 8 shows a furnace 70 with negative pressure-applying apparatus 71 by means of which unilateral stresses can be set up in mica by placing the mica in unilateral tension. The furnace 70 includes a refractory floor 72 and side walls 73 with resistance elements 74 embedded in the refractory which can have a suitable liner, as previously discussed. Resistance elements 75 are in a cover 76 which has a stepped recess 77 similar to a stepped recess 78 in the floor 72. The cover 76 is connected by means of a rotatable joint 79 to a threaded

shank 80 which extends through a threaded hole 81 in a block 82 held by supports 83 and 84 resting on beams 85. The support 84 merely contacts the corresponding beam 85 while the support 83 is pivotally attached to its corresponding beam 85 by a pivot 86. In this manner, when the cover 76 is above the furnace 70, the entire unit can be swung out of the way to enable free access to the furnace for batch loading. The threaded shank 80 is rotated through a bevel gear 87, a pinion bevel gear 88, and a motor and reducer combination 89.

The operation of this furnace is similar to those previously discussed except that when batch therein has been heated and melted for a desired period of time, the cover 76 is moved upwardly to place the batch in unilateral tension throughout a large part of the batch. The recesses 77 and 78 have coolant tubes 90 through which coolant can be supplied when the batch is ready to be cooled to solidify the batch adjacent the stepped recesses 77 and 78 and thereby enable the batch to adhere better to the floor 72 and to the cover 76 as it is raised.

The invention basically comprises means for suitably heat-treating a synthetic mica batch and slowly cooling the batch while applying pressure thereto. The pressure can be negative or positive and so applied as to establish uniaxial, biaxial, or multiaxial stresses in the mica to control crystal growth therein and to enable a larger amount of larger crystals to be formed.

Numerous modifications will be apparent particularly relating to apparatus for carrying out the principles of the invention. Such modifications can be made without departing from the scope of the invention as defined in the appended claims.

I claim:

1. A method of producing crystalline shapes from a synthetic mica material which comprises melting a batch of the material, slowly cooling the molten material through a range in which constituents thereof tend to crystallize by fractional crystallization, and moving an exterior portion of the batch in a direction having a component perpendicular to the body thereof to apply controlled, uniform pressure to the exterior portion of the material as it is cooled through said range, said pressure being different than that on at least one other exterior portion of the material whereby crystals tending to form will grow in a direction toward at least one exterior portion on which the pressure is less.

2. A method according to claim 1 wherein the pressure is increased as the mica cools.

3. A method according to claim 1 wherein the pressure is increased at an increasing rate.

4. In a method of producing synthetic mica which includes the steps of preparing a synthetic mica batch, melting the batch, and slowly cooling the molten batch through a crystal-forming range, the improvement which comprises supporting the batch with a first exterior portion thereof in a fixed position, and causing movement of a second opposed exterior portion of the batch substantially perpendicularly to at least one of said first and said second exterior portions as the molten batch is cooled through at least a substantial part of the crystal-forming range and while some of said batch remains liquid and crystals form and grow in said batch, whereby a force tending to cause interior movement within the liquid body is established and maintained and growth of said crystals is increased in at least one direction.

5. A method according to claim 4 wherein the movement of the second exterior portion is toward the first portion.

6. A method according to claim 4 wherein the movement of the second exterior portion is away from the first portion.

7. In a method of producing synthetic mica which includes the steps of preparing a synthetic mica batch, melting the batch, and slowly cooling the molten batch through a crystal-forming range, the improvement which

comprises supporting the batch with two planar exterior portions thereof in fixed, mutually perpendicular positions, and causing lateral movement, relative to the body of the batch, of substantially every exterior portion thereof which is opposed to one of the first two portions as the molten batch is cooled through at least a substantial portion of the crystal-forming range and while some of said batch remains liquid and some of said batch crystalizes, whereby growth of crystals is increased in at least one direction, the lateral movement having a component which is perpendicular to at least one of said first two portions.

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