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TREATMENT OF LAMINATED MATERIAL

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This invention relates to the treatment of material composed of elemental structural parts which are parallel or substantially parallel such as in a laminated or foliated material typified by mica, or whose elements are fibrillated or arranged in a similar manner and are composed of fibers or fibrous material such as in asbestos, palygorskite and the like. More particularly, the invention relates to the separation into its elements of such material, regardless of their water content and to the reconstitution of such elements, when indicated, in forms which are industrially useful.

The use of natural mica in electrical insulating applications is well established. The earliest usage of such mica for this purpose was probably in the form of single sheets or crystals and later pasted laminates of thin flakes arranged in overlapping fashion for tapes and other structures requiring larger areas than could be obtained from even the largest single crystals or flakes. Even more recently relatively small flakes or particles of mica have been reconstituted by paper making methods into a so-called mica mat or mica paper which has found usage in the electrical industry.

In order to reduce the natural mica blocks, books or flakes as received into a desirably thin state, various methods of splitting the material as received have been employed. For example, it is well known to hand split mica into thin flakes as by means of a knife. It is at once apparent that such a procedure is costly and inefficient. It is also well known to force jets of water at high velocity against mica particles, flakes, books or blocks in the hope that the jets will hit the flake or mica at a proper point or edgewise in order to cleave it. It will be quite apparent that in addition to the fortuitous cleavage which occurs, considerable grinding of the mica results. This method thus also results in the production of a considerable proportion of irregularly shaped mica fines as opposed to relatively large integral flakes. It is also well known to heat hydroxyl group containing natural mica whereby some of the water of constitution remaining therein is turned to steam and thereby puffing or expanding the material. Natural muscovite is the only material of the type herein referred to which has a sufficient amount of water of constitution to permit efficient puffing by heat as above. Usually the swollen or puffed mica is beat in machines similar to those used in paper production to break up the puffed material to yield a material which may be made into paper by the usual procedures. As pointed out above, this puffing process is only applicable to one form of natural mica, namely, muscovite, which contains the required water producing hydroxyl groups.

It is quite apparent that there is a definite need for a method of splitting or separating mica or other foliated material which will result in the production of relatively large flakes of material or flakes of material which are approximately, or as large as the flakes or crystals in the material being treated, at the same time subjecting the mica or similar material to a minimum degree of grinding action and which will produce maximum yields of flakes with high surface area to thickness ratios. It is also quite apparent that there is a definite need for means of so treating mica or similar material which does not lend itself because of its lack of water of constitution to other comminuting procedures such as puffing. Included are the natural phlogopites and other micas. Typical also

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of such materials are the more recently developed synthetic micaceous materials such as fluorophlogopite and the like which do not contain a sufficient amount of water of constitution for puffing procedures. At the same time, there is a need not only for efficiently reducing structures of the type described to their ultimate elements but a need for such a process which will eliminate the uncontrolled grinding and irregular comminuting action experienced in prior procedures.

A principal object, therefore, of this invention is to provide an improved method for splitting into its elemental material such as mica and the like or fibrous materials such as asbestos, etc., such method being entirely independent of the water content of the material.

Another object of the invention is to provide an improved method for obtaining flakes of micaceous material from the original blocks or books with a minimum reduction of the area of the elements.

A further object of the invention is to provide a method for reducing micaceous materials to their elemental structures which involves little or no grinding or irregular reduction of the material.

A still further object of the invention is to provide means for reconstituting such materials into useful forms by means of the same process through which they are reduced to their elemental structures or by independent processes.

Briefly, one aspect of the invention relates to an improved method of separating into its physical elemental structures material whose elements are substantially parallel which method comprises applying an adhesive to each side of the material and thereafter applying a peeling force to said material which is greater than the bonding force between the elements of the material. The procedure is repeated to obtain successively finer elements. This method can be applied to the production of large flakes or splittings or elements from material whose original elements are of such dimension. Likewise, it applies to the production of relatively smaller or finer elements from materials whose elements are smaller. It also relates to the continuous production of pasted tapes or similar structures from the elements so produced and to the preparation of paperlike materials or mats from such material particularly when the elements thereof are relatively small.

The reduction of the micaceous or similar material according to the present invention may be simply carried out. It is only necessary that an adhesive be applied to each side of the material after which a peeling force is applied to the sides of the material such that the bonding force between the elements of the material is overcome and the material peeled apart. For example, an adhesive coated tape such as that used for ordinary household purposes may be applied to each side of a flake and then be slowly peeled apart, each piece of tape carrying with it a reduced flake of the micaceous material. However, as a practical matter and for production purposes, it is preferable to use a system of two or more rotating members such as rolls or belts rotating in opposite directions to which the adhesive and the mica or other material to be cleaved or reduced is fed in well known manners. The micaceous or other structure is coated on each side by the adhesive material and as it passes through the nip of the rollers, belts and the like, a peeling force is applied to either side by the adhesive clinging to the rolls, etc. whereby the structure is peeled apart from either side, the unpeeled portion proceeding around the periphery of the roll, etc. to be once again coated with adhesive and split by the peeling action as it passes through the nip. This procedure can be repeated as long as desired, each passage through the nip of the system further reducing the material. The separated or peeled material, of course,

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remains on the roll or belt building up thereon in a layer or layers which are bound together by the adhesive material. While, as pointed out above, a single pair of rollers or belts may be used, it has often been found desirable to use arrangements of multiple rolls or belts, the mica and adhesive being fed into the system at any point and quickly transferred over the entire system. When the rolls or other rotating members are of different sizes, different portions of the rolls or rotating members will be presented at the nip at each rotation.

Any of a number of adhesives may be used in connection with the present invention including molasses, corn syrup, burnt sugar syrup and animal glues, as well as resins typified but not limited to by the thermosetting phenolics, polyesters, epoxy resins, silicone resins, butadiene resins, drying oils, furan resins, two-stage resins of various types as well as asphalts, tars, bitumens, the only requirement being that such adhesives, no matter what their origin, have a sufficient amount of tack or adhesiveness to facilitate delamination or reduction of the material. Other suitable adhesives will occur to those skilled in the art. It will be realized that temperature and humidity conditions will affect the adhesiveness of the various materials and such parameters may be varied in well known manners to insure the proper tack in the adhesive under any given conditions. The solvent content of the adhesives can also be varied to provide the proper tack under various conditions. A general rule for testing the proper tack of the adhesive is that when a portion of it is placed between the thumb and forefinger and the fingers separated, the material will string. In some instances where high temperature bonds or bonding material is desired, materials such as borax and other synthetic and naturally occurring glasses as well as synthetic glasses may be used as the adhesive material. Inorganic polymers such as silicates, polyphosphates and the like can also be used.

When the micaceous or other material being reduced is desired in its uncombined form or as such the adhesive material used for defoliating can be removed by applying thereto any well known solvent for the particular adhesive used. Thus, water can be used as a solvent for corn syrup which latter may be recovered from solution for further use. Likewise, various suitable organic solvents well known to those skilled in the art can be used for resinous materials. Likewise, where high temperature melting materials such as glasses and the like are used, subjection to high temperature as well as selective solvents may be used to free the material being separated.

In addition to separating the material described herein, the present process may also be used as an adjunct of the separating procedure to form or build up structures from the micaceous materials. Thus, mica tapes and the like can be formed by using a suitable resin as the adhesive material. After a desired thickness of resin bonded mica has been built up on the roll, the layer may be split in a helical fashion and removed from the roll in tape form. Likewise, cylinders of resin bonded material may be provided by simply building up the proper thickness of material on the roll and removing the cylinder so formed from the roll. Likewise, conically shaped structures can be made by using rolls of suitable shape or by adjusting the opening between the rolls to cause the desired selective build-up of material on the rolls. It will be understood, of course, that where resins are used, these may be cured in place on the rolls by usual well known procedures. Additionally, of course, it will be realized that where the material being reduced to its elements contains inherently relatively small sized elements, laminates may be made therefrom on the rolls themselves or, alternatively, the reduced material may be recovered by solvent or other well known methods and made into mat or paper by usual paper making techniques.

The following examples will illustrate the practice of

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the present invention, it being realized that such examples are to be taken as illustrative only of the invention and not limiting in any way.

Example 1

A thin layer of tacky, alcohol soluble phenolic resin was spread on a canvas strip (approximately 4 x 30 inches) and several grams of synthetic fluorophlogopite mica flakes (approximately 0.5 x 0.5 inch size) were scattered on the tacky surface. The strip of canvas was folded to bring the tacky mica bearing surfaces together and then pulled apart rapidly to produce a peeling action. The folding together and peeling action was repeated a number of times until most of the surface was coated with thin replicas of the original flakes. At this point, additional adhesive was applied to the mica coated canvas and the folding-peeling action was repeated. Finally, the canvas strip was immersed in alcohol to dissolve the resin and the highly delaminated flakes were recovered from the alcohol wash liquor. The recovered flakes were thoroughly washed with alcohol and then slurried in water and sheeted on a Büchner funnel to yield a mica paper.

Example 2

To mechanize the process of Example 1, similar synthetic fluorophlogopite flakes were sprinkled on a miniature set of differential size mill rolls which were coated with the same resin. The rolls were set 0.1 inch apart to prevent crushing or grinding of the flakes. The product was recovered in a manner similar to that of Example 1, that is, the adhesive containing the delaminated mica was doctored off the rolls and then dissolved in alcohol to free the mica. Again, an integral sheet of mica paper was prepared on a Büchner funnel from a water slurry.

Example 3

The process of Example 2 was repeated several times to include variables such as premixing adhesive and mica chips and substitution of molasses and caramelized sugar for the tacky phenolic resin. The latter substitutions permitted the use of water washing in place of alcohol. A number of integral sheets were prepared.

Example 4

A pair of cork rolls four inches in diameter and six inches long were constructed and one of them was connected to a variable speed drive. The second roll was friction driven by the first. Contact between the two rolls was maintained by a light spring loading. A dispersion of 5 grams of coarse mica chips in 50 grams of blackstrap molasses was added to the rolls (70 r.p.m.) over a 24-minute period of time. The product was recovered by washing the rolls in water which simultaneously dissolved the molasses.

Example 5

The two rolls described in Example 4 were separated approximately 2.5 inches and a third roll consisting of a quart jar was placed in the saddle between the two. The third roll had the effect of increasing the total surface area and doubling the number of nips (lines of contact between rolls). Using a drive roll speed of 80 r.p.m., 20 grams of mica chips and 124 grams of blackstrap molasses were alternately applied to the delaminator over a period of 110 minutes. The procedure consisted of applying a few drops of adhesive, waiting for a short time until it was distributed over the rolls and then sprinkling a small amount of mica chips on the tacky surface. The mica delaminated, covered the tacky surface and more adhesive was added to repeat the cycle. The delaminated mica was recovered by water washing and filtration of the wash solution through a 325 mesh sizing screen. The washed product could be sheeted in a Büchner funnel from a water dispersion.

Example 6

A five-roll delaminator was constructed. The 3 bottom

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rolls were washing machine wringer rolls which were 2.5-3.0 inches in diameter and 12 inches long. These rolls were arranged parallel to each other with centers 8.5 inches apart. Two 8 inch by 12 inch rubber coated rolls were cradled between the bottom wringer rolls. One of the bottom rolls was driven, the other four rolls were friction driven. The weight of the 8-inch rolls served to maintain uniform pressure between all of the rolls. Data from one run on this equipment is given below:

Adhesive: 419 grams of corn syrup applied as needed by draining from a spatula.

Mica Feed: 151 grams of synthetic fluorophlogopite (-2 +4 size—U.S. Standard screen size) sprinkled on by hand as needed over a period of 50 min.

Roll Speed: Bottom drive roll rotated at 175 r.p.m.

Results: The product was recovered by flushing the system with water and wet screening through a U.S. Standard 16 screen and retaining product on a 325 screen. Nine grams of oversize material (+16) and a negligible amount of undersize (-325) were obtained. Paper was prepared from a number of screen fractions. The -16 +230 fraction yielded paper with a tensile strength of 1200 p.s.i. and a dielectric strength of 500 volts/mil. An 8 mil (0.008 inch) thick sheet was used for testing.

Example 7

Another run on the equipment described in Example 6 was made in which the ratio of mica chip to corn syrup was 1:3.8. Several hand sheets were prepared from the -16 +325 fraction of the product. Tensile strengths for 1-2 mil sheets varied from 3000-4600 p.s.i. and dielectric strength varied from 700-950 volts/mil.

Example 8

In another experiment on the previously described 5-roll system, the adhesive was automatically applied in a circular pattern over the center of the rolls by a rotating (motor driven) system. The mica chips were fed by means of a vibratory chute equipped with an interval timer. A mica to corn syrup ratio of 0.9:1.0 was used. Five hundred grams of mica was delaminated in 85 minutes with a drive roll speed of 175 r.p.m. The -16 +325 product yielded paper which tested in excess of 3000 p.s.i.

Example 9

A seven roll delaminator was constructed consisting of four washing machine wringer rolls on the bottom, two 8-inch and one 10-inch rolls on top. All of the bottom rolls were arranged to be driven, the top rolls were turned by idling on the bottom driven rolls. Several perforated copper tubes were arranged above and parallel to the large top rolls. The copper tubes were connected through a series of valves to a gear pump for metering adhesive. This arrangement permitted application of adhesive to each or all of the top rolls as desired. A vibratory chute feeder connected to an interval timer was arranged to scatter mica chips at the nip of two end rolls. Using this system a large number of runs were made in which the corn syrup adhesive to mica ratio varied from 5:1 to 0.5:1 and roll speeds (drive roll) varied from 100 to 250 r.p.m., the feed rate of mica varied from 50 grams per hour to 1000 grams per hour. The products were generally recovered by washing the rolls with water and screening the wash water. For control purposes, sheets of paper were prepared by air drying a gravity settled slurry on various sizing screens. The fractions passing through a -16 standard screen and retained on a 325 screen yielded tensile strengths from several hundred p.s.i. to 5000 p.s.i. or higher. Selected screen fractions, for example, through 30 mesh and retained on 70 mesh, gave tensile strengths approaching 10,000 p.s.i. Damp pressing and/or steaming raised the tensile strength of all samples. When the high tensile strength sheets were damp pressed between steam heated platens, tensile strengths as high as 15,000 p.s.i. were obtained. Average paper prepared by gravity settling and drying on screens has an apparent density of 1.4

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to 1.5, a dielectric strength of approximately 500-600 volts per mil and a tensile strength of 2000-4000. Higher tensile paper from selected fractions, or prepared by steam pressing, has a density of 1.8-1.9 and dielectric strength of 1000 volts per mil or higher in thicknesses of 1-5 mils.

Example 10

When the rolls such as those described in previous examples are slightly out of parallel, there is a tendency for the delaminated mica-adhesive cake to migrate or extruded toward one end of the rolls. This material may be doctored off the ends of the rolls and the mica may be recovered by the same technique of dissolving the adhesive and screening or sieving the product out of the water. Several narrow rolls (solid rubber tires) were arranged with an international canted arrangement to permit continuous delamination and extrusion of the delaminated product. The recovered product made satisfactory paper.

Example 11

When both mica chips and adhesive are fed at one end of a roll system such as described in Example 9, there is a tendency for the delaminated material to migrate toward the opposite end of the set of rolls. The migration can be encouraged by removing delaminated mica-adhesive cake from the end roll. This arrangement lends itself to a continuous process. The adhesive-mica cake doctored off of the end roll makes satisfactory paper.

The previous examples were based on synthetic fluorophlogopite mica. To show the general applicability of the process, other micas were delaminated such as Bengal ruby muscovite, native (North Carolina and New Hampshire) muscovite, Madagascar and Brazilian phlogopite.

Example 12

Using the 5-roll delaminator described in Example 6, 110 grams of Bengal ruby mica (1-2 inch square pieces, 5-10 mils thick) and 458 grams of corn syrup were added to the rolls over a 50-minute period. The drive roll was initially run at 130 r.p.m. and was cut down to 60 r.p.m. near the end of the run. The product was recovered as previously described. A portion of the -16 +325 screen fraction yielded mica paper with a tensile strength of 1080 p.s.i.

Example 13

Twenty grams of the same mica used in Example 12 was delaminated on the same equipment in a period of 15 minutes with a drive roll speed of 175 r.p.m. The quantity of adhesive used was not measured. The -16 +325 fraction from this run yielded mica paper with a tensile strength of 4200 p.s.i.

Example 14

Twenty grams of fired (swollen by brief heating at elevated temperature) tube spacer muscovite scrap was delaminated under the same conditions as Example 13 except the corn syrup was diluted with an equal volume of water. The -16 +325 fraction yielded paper with a tensile strength of 2900 p.s.i.

Example 15

The equipment described in Example 9 was used to delaminate Madagascar amber (phlogopite) mica. The mica was cut with scissors to approximately 1 inch square with an average thickness of 5-7 mils. The adhesive was an industrial corn syrup with a viscosity of 2200 centipoises. Six hundred grams of adhesive were fed by gear pump over a 57 minute period of time. The mica, 285 grams, was hand fed because the vibratory feeder was not calibrated for this size of material. The speed of the ten-inch idling roll was 40 r.p.m. The yield of -16 +325 fraction from this run was 140 grams, the remainder was primarily oversize which could be re-run. The paper from this run had a tensile strength of 2900 p.s.i.

Example 16

Native muscovite was delaminated using the same equipment as described in Example 9. Using a drive roll speed of 136 r.p.m., 343 grams of mica and 395 grams of corn syrup were fed to the system over a 70 minute period of time. Paper produced from this delamination product tested 1730 p.s.i. and 700 volts per mil.

To further demonstrate the versatility of the system, fibrous materials were pulled apart or shredded using the same general technique.

Example 17

Small lumps or blocks of chrysotile asbestos were fed to a three-roll delaminator using corn syrup as the adhesive. The material was rapidly shredded to yield a fibrous mat-like composite on the rolls. The asbestos fibers were recovered by washing in water. The washed material was dispersed in water and after permitting a short settling time to eliminate incompletely defibrillated material, the supernatant slurry was filtered through a Büchner funnel. A strong asbestos paper was formed.

Example 18—Palygorskite

Ten grams of Brazilian palygorskite, a mineral commonly called mountain leather or cork, were defibrillated on the same rolls used in Example 17. The product was recovered by dissolving the adhesive in water and filtering the suspended fibrous material on a Büchner funnel. A 4-mil and a 7-mil thick sheet were made in this manner. The samples had tensile strength of 250 p.s.i. and dielectric strength of 300–350 volts per mil.

Throughout the previous examples, the principal adhesive used was corn syrup. In general, this type of adhesive is particularly convenient to work with since it contains very little contaminating material which cannot be removed conveniently with a simple water wash treatment. In addition to this variety of water-soluble adhesive, molasses (refined and crude), various invert sugars and caramelized cane sugars have been used.

Non-water soluble adhesives were also used. An example using an alcohol soluble phenolic resin was previously listed. In addition, pitches, tars and a tacky variety of heavy residual refinery oil satisfactorily delaminated the mica. Hydrocarbon solvents were required to remove these adhesives prior to making paper.

Several inorganic adhesives were also used, including thickened phosphoric acid based cements. A somewhat different example is given below.

Example 19

A small delaminator was constructed from three short lengths of stainless steel pipes. Several Bunsen burner flames were played on the turning rolls until they were sufficiently hot (500–600° C.) to dehydrate and melt boric acid. Powdered boric acid was then dusted on the rolls and within a few seconds it became quite tacky and stringy. Mica chips were then added to the rolls and the process of adding powdered boric acid and mica chips was repeated a number of times. The product was recovered by soaking the rolls in water to dissolve the boric acid glass. A satisfactory sheet of mica paper was produced from mica delaminated by this system.

All of the previous examples were directed toward delamination or defibrillation of material to reduce it to the proper size for the production of a paper or mat type product through the use of paper making techniques. However, it is desirable for some purposes to produce a product which is actually a mica paper type product impregnated with insulating resins. The adhesive delamination process can be used to advantage to produce novel laminates and laminated structures. The following examples illustrate some of the various techniques and materials which have been used.

Example 20

The three roll delaminator described in Example 5 was used to prepare a novel mica-phenolic laminate. The rolls were covered with a sacrifice layer of 2 mil polyethylene film prior to the delamination step. Twenty grams of mica chips and 90 grams of tacky phenolic resin were added to the rotating (92 r.p.m.) rolls over a period of 55 minutes. The effect was to delaminate the mica chips and rebond them in a laminar structure with the uncured phenolic resin. At the conclusion of the run, the delaminated-re laminated layers were removed from the rolls by slitting and peeling off of the sacrifice layer of polyethylene film. On chilling in a Dry Ice chest, the phenolic laminate became stiff and the polyethylene backer could be readily removed. Three, 4 x 4 inch pieces of the phenolic-mica composite were stacked together and cured under pressure in a steam heated press to form a useful structure.

Example 21

A set of rolls was constructed from two washing machine wringer rolls spaced about six inches apart and a top 8 inch diameter floating roll. The rolls were covered with polyethylene film to facilitate removal of the material and to prevent solvent action on the rolls by various resins and solvents. The film was wrapped around the rolls and the end was held in place with masking tape. A dodecylsuccinic anhydride catalyzed epoxy resin (Epon 828—300 grams) was used as adhesive. The adhesive and 156 grams of synthetic fluorophlogopite chips were applied to the rotating rolls over a period of 75 minutes. The roll speed was somewhat higher than usual because the resin was of lower viscosity and less tacky than previous adhesives. The effect of increasing roll speed tends to compensate for lower viscosity adhesives very conveniently. The polyethylene backed laminate was removed from the rolls, separated from the backer as in the previous example and pieces were stacked and cured. The laminate could be sawed or cut with a paper knife type of sample cutter. Thin samples were flexible.

Example 22

The equipment described in Example 21 was used to produce a silicone bonded laminate. A pressure sensitive silicone adhesive was diluted with toluene to a desirable tacky state (strings form when a small sample is pinched between the forefinger and thumb and the fingers are separated) and used to delaminate 300 grams of synthetic fluorophlogopite mica. The product was cut off the large roll in tape form. This product was not cured but the solvent was evaporated slowly to yield a flexible, somewhat elastic tape which contained 40% silicone resin.

Example 23

A concentrated solution of shellac in alcohol was used with the same general procedure as in the previous example. After evaporation of the solvent, several plies of the material was hot pressed to yield a shellac bonded mica laminate which was reasonably flexible and cut readily with a paper cutter. An 0.058 inch thick sample had a power factor of 0.0122 and a dielectric constant of 2.80 at room temperature.

Example 24

A glass fiber reinforced mica-epoxy composite structure was prepared using the same equipment as in the previous example. In this case, the film protected roll was rendered tacky by the addition of a small amount of polyamide catalyzed epoxy resin (Epon 828) and the rolls were wrapped reel fashion with glass roving. The glass was applied approximately one layer thick and then mica chips and adhesive were alternately added to produce a laminar mica-epoxy structure. At the conclusion of the run, the product was recovered as a one inch wide tape by cutting the roll coverings off on a bias. The effect

of the bias cutting was to yield a tape composed of a layer of polyethylene, on top of which was a layer of diagonal glass fibers embedded in epoxy resin and then a layer of laminar mica particles embedded in epoxy resin. The glass fibers were approximately two inches long running from one edge of the one inch wide tape to the other at an approximate angle of 30° with the direction of the tape. The glass reinforcement contributed considerably to the tensile strength of the uncured tape. The polyethylene was removed from some of the tape and the tape was wrapped around an electrical conductor and cured.

Example 25

A liquid rubber formulation composed of liquid synthetic rubber (Hycar), benzothiazyl disulfide, sulfur, zinc oxide and stearic acid was used to delaminate mica chips on the same equipment described in Example 21. The end product laminate contained 295 grams of the rubber and 180 grams of mica. It was cured at 100 p.s.i. and 150° C. to yield a 0.051 inch thick laminate with a room temperature power factor of 0.123 and a dielectric constant of 0.324.

Example 26

Extremely thin glass flakes are prepared by blowing glass bubbles and smashing them or by some form of this technique. These flakes have been used as fillers for various resinous laminates. The standard procedure of incorporating these flakes in laminates frequently results in several flakes being coated with resin in such a manner that the glass to glass space between agglomerated flakes is not wet with resin. This has an undesirable effect on finished laminates since these points can absorb water vapor or introduce discontinuities in electrical properties. The adhesive roll technique was used to produce a glass flake mica laminate by mixing the glass and mica chips before feeding them to the adhesive coated rolls. The mica flakes were delaminated and the glass flakes which would normally tend to agglomerate were pulled apart and individually embedded in resin. A sample of laminate prepared from this combination with an epoxy adhesive had the following properties. A 25 mil sheet had a power factor of 0.012 and a dielectric constant of 2.36 at room temperature. A freshly cut edge was immersed in a dye used to determine wicking of polyester glass laminates and no wicking was apparent. It will be realized, of course, that the glass flakes can be used alone to produce glass flake-adhesive laminates in which inter-flakes interstices are entirely filled with adhesive.

Example 27

A non-curing laminate was prepared by delaminating 66 grams of mica with 47 grams of tall oil pitch. The pitch was reduced in viscosity by directing heat lamps on the rolls. The product was flexible and could be built up by pressing several layers together in a heated press.

Example 28

An aluminum phosphate type inorganic cement was thickened by partial dehydration until it became slightly tacky. This was then used to make a mica laminate on the adhesive rolls. The final product was cured by heating in a press at 150° C. and finally by short time direct firing with a Bunsen flame. The product could be cut with a paper knife type sample cutter with little edge damage. The strips were reasonably flexible.

Example 29

A set of two stainless steel rolls were constructed from six lengths of 3.0 to 3.5 inch tubing. Mandrels were constructed to slide on stainless steel hollow shafting to act as end-plates for the tubular rolls. A high temperature furnace was constructed to enclose the two rolls. One of the rolls was driven by a variable drive, the other was maintained against the driven roll by spring loading.

Bearings and spring loading device was arranged outside of the furnace and provision was made to water cool the bearings. Silicon carbide resistance heaters were used to heat and hold temperatures in the furnace. A top opening was provided to feed material to the rolls. Using this system, powdered soft glass was added to the turning rolls and the temperature was raised until the glass became tacky and stringy. At this point synthetic fluorophlogopite mica chips were added and rapidly delaminated. The process of adding powdered glass and mica chips was repeated many times over a period 30-40 minutes. The heaters were shut off and the rolls were cooled by blowing air through the hollow shafts which interconnected with the hollow rolls. When the temperature cooled to the point where the rolls could be handled with gloves, the mandrels on one end of the rolls were removed and the roll bodies (stainless steel tubing) were slid out one of the dissembled furnace. Due to differential shrinking of the steel and the glass bonded mica structure, the glass bonded laminates could be removed readily. One of the laminates was removed as a cylinder, the other was cut parallel to the roll on opposite sides and removed as two half cylinders. The laminar mica filled glass composite was flattened into a sheet by soaking in a 500-600° oven until the glass binder softened. The composite is somewhat flexible.

Example 30

Using the equipment described in Example 29, a number of cylindrical glass-mica laminates were prepared using low melting lead glasses, a cobalt glass and Pyrex glass.

Example 31

Two tapered steel rolls were constructed. The angle of taper was the same on both rolls but they were approximately 0.5 inch different in diameter. These rolls were mounted parallel to each other (one driven, one idling against the driven roll) with the taper on the one opposite that of the other. A layer of paper held by adhesive tape was placed on each as a form of mold release. Mica chips and catalyzed epoxy resin were fed to the rotating rolls to build up laminar cone shapes. After approximately 0.125 inch of laminate was formed, the rolls were separated and a very narrow (0.75 inch wide) roll was applied to the large end of the driven tapered roll. Mica and more adhesive were added at the nip of this roll combination to build a thickened reinforcing band. Both laminar cones were cured with heat lamps and removed from the steel rolls. The cones could be flexed without breaking.

Most of the previous examples tended to produce relatively small and very thick flakes of mica which either could be reclaimed for paper making or left as an integral part of bonded laminates. For some purposes it is desirable to produce a pasted mica type sheet in which large (one to two or even three inch square pieces and perhaps 0.001 inch thick) thin sheets of mica are randomly laid out several plies thick and bonded with various adhesives. It is standard procedure in making a sheet of this type to start with hand split mica. I have found that I can eliminate the original hand splitting and simultaneously split books of mica, lay them up in a random shingle type fashion and cement them together with one operation.

Example 32

An 8 inch by twelve inch hard wood roll was arranged on a tilting track so that it could be driven by a variable drive. A ten inch by twelve inch rubber coated roll was placed on the inclined track above the drive roll in a manner such that it could be friction driven by the drive roll. Contact pressure between the rolls could be varied by increasing or decreasing the angle of incline of the track. The idling roll could also be filled with water or other media to increase its weight and hence the contact pressure. Both rolls were covered with polyvinyl alcohol

film to facilitate later removal of the laminated mica sheets. A polyamide catalyzed liquid epoxy resin was used as adhesive. The mica feed was muscovite mica punching scrap which measured approximately 1 x 2 x 0.006 inches. One half inch holes had been punched in the middle of this scrap. The drive roll was turned at 100 r.p.m. and punchings and resin were fed alternately to the system over a 30 minute period of time. The mica was fed at a rate sufficiently fast to prevent marked reduction in size of the scrap, the principal effect was to reduce the punchings to approximately 0.0005 inch in thickness. The laminate was removed from the rolls in sheets which were 12 inches wide and 3.14 times their respective diameters of eight and ten inches. The pasted sheets were cured for two hours at 125° C. in an oven without applied pressure. The polyvinyl alcohol sacrifice layer was removed and the sheets were evaluated. The resin content was 38%, the dielectric strength varied from 3-400 volts per mil for thicknesses from 30-40 mils.

Example 33

The process of Example 32 was repeated using similar sized pieces of regular Madagascar phlogopite with the same type of product resulting.

From the above it will be seen that there is provided by the present invention improved means for defoliating or defibrillating materials such as mica and fibrous materials which is entirely independent of moisture content of the material and which produces reduced elements far superior to those provided by former methods with minimum of grinding or irregular communication and wasting of material. By the present invention there is also provided means for producing useful structures from such delaminated or defoliated material which process is integral with that used for reduction.

What I claim as new and desire to secure by Letters Patent of the United States is:

1. The method of preparing composites from materials having laminar, fibrillated and foliated elements which comprises repetitively passing said materials through the nip between the surfaces of rotating members, said members having adhesive applied to the surfaces thereof and being so constructed and arranged that different portions of the surfaces of said members are presented at the nip

between said members at successive rotations whereby said materials are subjected to peeling forces which separate the materials into their elements, said elements with the adhesive forming a composite layer on the surfaces of said members.

2. The method of separating into its physical elements materials whose elements are substantially parallel which comprises repetitively passing said material through the nip between the surfaces of rotating members, said members having adhesive applied to the surfaces thereof and being so constructed and arranged that different portions of the surfaces of said members are presented at the nip between said members at successive rotations whereby said material is subjected to peeling forces which separate said elements.

3. The method of separating mica which comprises repetitively passing said mica through the nip between the surfaces of rotating members, said members having adhesive applied to the surfaces thereof and being so constructed and arranged that different portions of the surfaces of said members are presented at the nip between said members at successive rotations, whereby said mica is subjected to peeling forces which separate said mica.

4. The method of preparing paper for laminated and fibrillated materials which comprises repetitively passing said materials through the nip between the surfaces of rotating members, said members having adhesive applied to the surfaces thereof and being so constructed and arranged that different portions of the surfaces of said members are presented at the nip between said members at successive rotations whereby said material is subjected to peeling forces which separate the elements of said materials, removing said adhesive from the separated elements and forming the separated elements into paper.

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