May 10, 1949. W. H. SMYERS 2,469,416

INSULATED CONDUCTOR

Filed Dec. 31, 1941 2 Sheets-Sheet 1

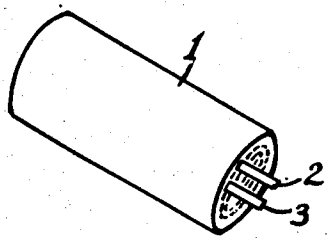


FIG.—1

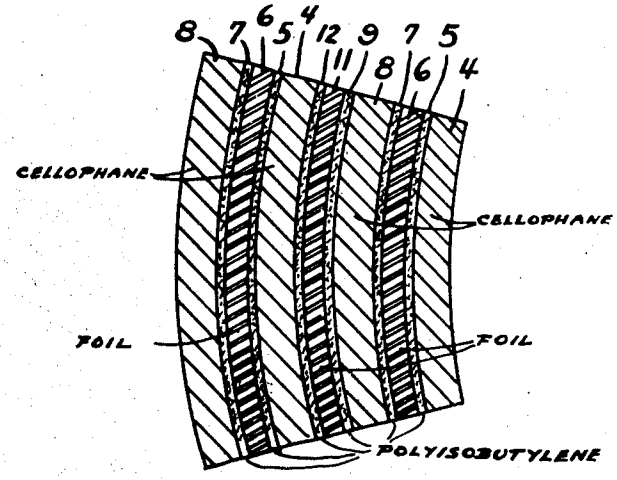


FIG.—2

William H. Smyers Inventor

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May 10, 1949. W. H. SMYERS 2,469,416

INSULATED CONDUCTOR

Filed Dec. 31, 1941 2 Sheets-Sheet 2

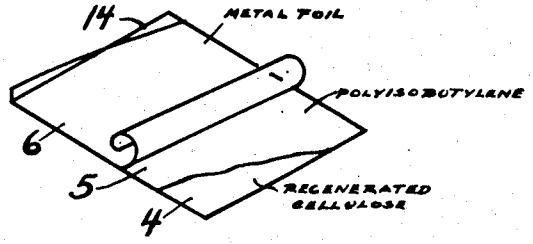


FIG.-3

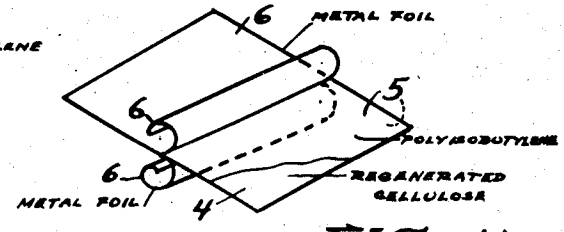


FIG.-4

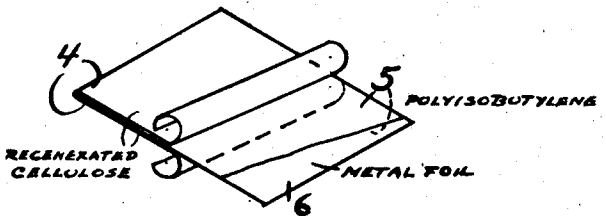


FIG.-5

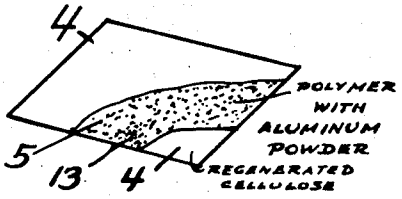


FIG.-6

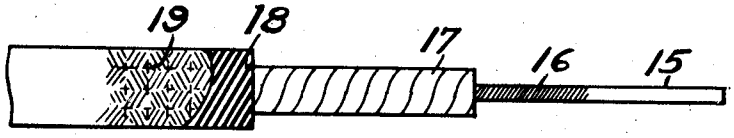


FIG.-7

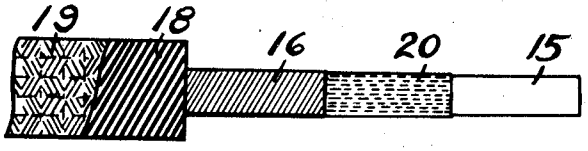


FIG.-8

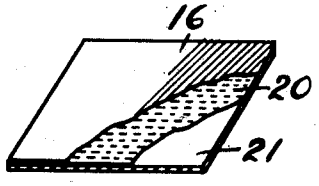


FIG.-9

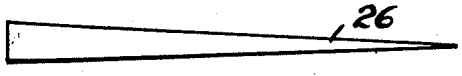


FIG.-11

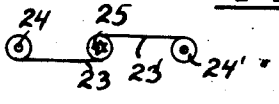


FIG.-10

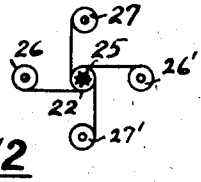


FIG.-12

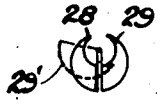


FIG.-13

William H. Smyers Inventor

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Patented May 10, 1949

2,469,416

UNITED STATES PATENT OFFICE

2,469,416

INSULATED CONDUCTOR

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Application December 31, 1941, Serial No. 425,031

4 Claims. (Cl. 174—120)

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This invention relates to combinations of homogeneous, smooth surface, cellulosic material, such as cellophane, attached to a metal surface by polyisobutylene as an adhesive; relates particularly to metal surfaces protected by cellophane attached to the metal surface by polyisobutylene; and relates especially to metallic, electrical conductors having a covering thereon of cellophane attached thereto by polyisobutylene.

This application is a continuation in part of my application, Serial No. 670,730, filed May 12, 1933, now Patent No. 2,226,589, patented December 31, 1940; my application, Serial No. 170,048, filed October 20, 1937, now Patent No. 2,226,590, patented December 31, 1940; my application Serial No. 704,747, filed December 30, 1933, now Patent No. 2,280,860, patented April 28, 1942; and my application, Serial No. 273,467, filed May 13, 1939, now Patent No. 2,300,072, patented October 27, 1942.

In the making of laminar sheets of metal protected with cellophane, and especially in the making of insulated, electrical conductors for various purposes; and especially in the making of rolled foil condensers for electrical service, it has been customary to roll up alternate layers of paper, preferably treated with paraffin, and foil, usually tin or aluminum foil, until a sufficient area of foil has been rolled to provide the desired electrostatic capacity. The electrostatic capacity per unit area of foil surface is a function of the special separation between the respective foil plates of the condenser, and a function of the dielectric constant of the interposed insulating material. Thus, the thinner the insulation between the foil plates of the condenser, the greater the capacitance of a given area of foil electrode surface, and a condenser may be smaller and contain less material for a given capacitance when the insulating material is thin than when it is thick. However, a primary requirement in any condenser is that the breakdown voltage of the insulating material interposed between the plates must be sufficiently high to withstand indefinitely the peak voltage applied between the plates of the condenser. Thus, strict limits in thinness of the permissible condenser dielectric occur which prevent more than very slight reduction of the spacing between the foil plates of the condenser.

But the breakdown voltage in any dielectric is a function of the distribution of the potential gradient. If the interposed dielectric is homogeneous, the electrostatic gradient between the

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plates occurs as smooth, even curves, with no foci or points of concentration of electrostatic forces.

To the present, however, it has not been possible to find homogeneous materials in thin sheets suitable for condenser dielectric. Paper is a matted body of fibres, with interspersed air between and around the cellulose fibres. Accordingly, the surface of every fibre is a focal point for a concentration of potential gradient which builds up quite rapidly, and readily reaches values sufficiently high to ionize the adjacent air, resulting in an immediate electrical breakdown and puncture of the dielectric material, whereupon the condenser becomes completely useless since the charring of the paper incident to the first passage of current, whether spark or arc, develops a conducting spot through the insulation. The introduction of a thorough impregnation of the paper by paraffin modifies the distribution of the electrostatic gradient in a favorable direction, and substantially increases the breakdown voltage of the paper, both by removal of the readily ionizable air, and by reduction of the voltage gradient concentration, since there is less difference in the dielectric constant of paraffin compared to cellulose fibres than there is between cellulose fibres and air. Thus a condenser made with paraffined paper for the dielectric will withstand greatly increased voltage which may be two or three times the permissible voltage with a plain paper and foil condenser. However, even paraffin is not satisfactory, since it does have a dielectric constant different from cellulose and there are residual foci in the dielectric gradient between the plates.

A desirable dielectric for rolled condensers is regenerated cellulose, known under the trade name of "cellophane," but to the present this has not been usable, mainly because of the difficulty of assembling the condenser. Cellophane is a material to which relatively few substances will adhere, and accordingly very great difficulty is experienced in rolling together plain cellophane and foil, since the two will not adhere, will not stay rolled, and the foil does not stay centered in the sheet of cellophane. Furthermore, in view of the easy-tearing character both of cellophane and of foil, especially the latter, it is not possible to roll the condenser under sufficient tension to hold the various parts together. Also, air pockets occur between the cellophane and foil which serve as foci for concentration of the voltage gradient between the plates.

These considerations apply equally to electrical insulation generally, for which purpose cello-

phane is a highly desirable insulating material if its limitations can be overcome. Thus, it is convenient to prepare an insulated, electrical conductor by wrapping the conductor with successive layers of cellophane, the first layer being attached to the conductor by an interpositioned film of polyisobutylene, the successive additional layers required to make up a sufficient thickness of insulating material, being attached to each other by successive interposed films of polyisobutylene (Vistanex polybutene). This structure is particularly advantageous for conductors which must be positioned closely adjacent to other conductors having different applied voltages thereon since such an insulating covering has, by virtue of its relatively very high homogeneity and good uniformity of dielectric constant, a higher breakdown voltage per unit of thickness than is obtainable with any other type of insulation. This form of insulation is also particularly advantageous for the making of shielded conductors, such as for radio purposes and for the making of condenser bushings for the carrying conductors through metal cases, such as tranformer leads, condenser leads, radio circuit leads and the like, since the voltage gradient distributing metallic layers which form the plates in the condenser bushing are readily distributed at the appropriate locations in the body of the insulation, and in the case of shielded conductors, the layer of foil or sheet metal which forms the shield is readily interposed at an appropriate point in the sequence of layers of cellophane wrapped around the primary conductor.

Likewise, the structure is highly advantageous for the protection of oxide coatings upon metal, especially oxide coated conductors to which the film of polybutylene adheres with great tenacity and to which the layer of cellophane is very firmly attached by the film of polyisobutylene adhering to the oxide-coated metal. Likewise the structure is highly advantageous for the protection of a variable condenser of extreme range by a combination of sheets of cellophane as insulators with sheets of metal foil as condenser plates carried upon three rollers, the respective foils of the pair being attached to one roller upon which they may be rolled up in interpositioned relationship, the other ends being attached to other rollers onto which they can be transferred separately. In fact, the structure is highly advantageous for the production of any laminated product of metal and cellophane attached to each other by an interposed film of polyisobutylene, the combination being useful for a wide range of uses in addition to the electrical condensers and electrical conductors above mentioned, including such uses as impervious wrapping materials which are resistant both to aqueous liquids and to non-aqueous liquids and vapors for such purposes as wrapping comestibles and food stuffs generally for maintaining the original condition of moistness or for preserving them against attacks of vermin, moisture or other deteriorating influences; or for the protection of metal structures generally.

The present invention makes use of the new and unexpected fact that a polyisobutylene having a molecular weight above about 1,000 and possibly as high as 300,000 or higher but preferably lying between about 3,000 and 25,000 in some cases and between about 100,000 and 200,000 in other cases, is strongly adhesive both to cellophane and to metal foil. Intermediate polymers may of course also be used. Accordingly, the con-

denser of the present invention utilizes homogeneous layers of cellophane coated on one or both sides with films of adhesive polyisobutylene and layers of metal foil, bonded to the layers of cellophane by the adhesive effect of the polyisobutylene.

Thus the structure of the invention, including the laminar structure generally consisting of cellophane, polyisobutylene film and metal base, particularly in the form of electrical conductors generally, as well as electrostatic condensers of the invention, consists of plates formed of metal foil, between which are interposed one or more homogenous layers of cellophane, coated with thin homogeneous layers of polyisobutylene. The regenerated cellulose forming the cellophane layer is wholly homogeneous, and accordingly, there are no focal points for concentrating the electrostatic gradient between the boundary planes of the cellophane sheet; similarly, the layer of polyisobutylene also is homogeneous, and accordingly there are no focal points for the electrostatic gradient in the polyisobutylene. The fact that both the polyisobutylene and regenerated cellulose are amorphous and non-crystalline makes both completely homogeneous, whereas crystalline materials have variations in voltage gradient at the surfaces of the crystals. Likewise, the boundary surfaces of both layers are smooth plane surfaces, and accordingly while there is a change in the slope of the potential gradient at the interface, there are no focal points and accordingly no undue concentrations of voltage gradient. Also, the dielectric constants of cellophane and of polyisobutylene are so nearly the same that there is very little change in the slope of the voltage gradient even at the interface.

Thus the structure of the invention consists of a combination of one or a plurality of sheets of cellophane attached by a film of polyisobutylene to a metal surface, the sheets of cellophane, if several are used, being attached to each other by additional interposed films of polyisobutylene, especially in combination with electrical conductors, both heavy metal strands and metal foil, such as is found in metal foil structures of the type of metal foil condenser plates with interposed dielectric material in the form of homogeous layers of cellophane and polyisobutylene, the whole being if desired rolled into a cylindrical condenser, or folded into a flat condenser, or otherwise arranged according to the customary and usual procedure.

Thus an object of the invention is to combine together sheets of cellophane with metal structures, such as foil or heavier metal objects, especially electrical conductors, to yield protective and insulating sheets, and protection in insulating laminar coverings in which the high homogeneity of the cellophane is utilized for strength, resistance to corrosive and other harmful influences and when used with electrical conductors, to avoid voltage gradient focal points in the dielectric of an electrostatic condenser and to insulate the plates of the condenser with a material of high dielectric constant, high breakdown strength, and free from focal points of voltage gradient concentration, utilizing homogeneous layers of regenerated cellulose and polymer having smooth boundary planes between the condenser electrodes.

Other objects and details of the invention will be apparent from the following description when read in connection with the accompanying drawing wherein

Fig. 1 is a view in perspective of a rolled condenser according to the invention, and

Fig. 2 is a sectional view of a portion of a condenser constructed according to Fig. 1.

Figs. 3, 4, 5 and 6 are views in perspective of laminated products with parts turned back or cut away to show the construction, comprising various combinations of sheets of regenerated cellulose and metal foil, or metal powder, using polyisobutylene to bond the metal surface to the cellulosic surface; Fig. 3 shows 1 sheet of each, Fig. 4 two of metal foil and 1 of regenerated cellulose, Fig. 5 the reverse of the product in Fig. 4, and Fig. 6 metal powder between two sheets of regenerated cellulose.

Figs. 7 and 8 are longitudinal sections of electric cables embodying different features of the invention.

Fig. 9 is a perspective of a sheet of coated metal with parts of the two coating layers cut away.

Fig. 10 is a cross section through one winding roll and two supply rolls of a continuously variable electrostatic condenser.

Fig. 11 is an outline of a metal foil strip such as used in the condenser of Fig. 10.

Fig. 12 is a condenser similar to that of Fig. 10 but having four supply rolls instead of two.

Fig. 13 is a cross section of a variable air condenser.

The polyisobutylene may be prepared as follows:

In the processing of crude oil to produce extra quantities of gasoline boiling range hydrocarbons by cracking the crude oil or fractions boiling higher than gasoline, e. g., gas oil, kerosene, lubricating oil, etc. at elevated temperatures, there is produced about 15% on the weight of crude oil charged, of gaseous materials consisting of hydrogen, methane and its gaseous homologues, ethylene and its gaseous homologues, etc., including substantial quantities of isobutylene. It is found that isobutylene when condensed by cooling to a low temperature, below —10° C., e. g., —40° C., —80° C., or —100° C., diluted with a diluent-refrigerant such as liquefied propane, butane, ethylene, etc., and treated with an active halide Friedel-Crafts catalyst, e. g., boron trifluoride, polymerizes into a substantially colorless, high molecular weight hydrocarbon material of relatively very low unsaturation compared to the original isobutylene.

After the polymerization at the low temperature is complete, the polymeric material is separated from the diluent-refrigerant and other materials, and desirably purified by a careful washing and kneading operation. The material usually is used in dissolved form, and it is convenient at this stage to dissolve it in solvent and either settle or filter the solution, in order to remove any fragments of metal or other foreign material which may have entered the polymer in the course of washing and kneading operations.

This polymer appears to be a branched, linear chain hydrocarbon which is substantially saturated, having an iodine number usually less than 5 and often as low as or lower than 1, and is readily made with a molecular weight ranging from about 1,000 up to 300,000 or above, according to the purity of the isobutylene, and the lowness of the temperature at which polymerization occurs; several commercial forms are now marked, namely; about 15,000; 80,000; and about 150,000, M. W. A desirable material for the structure of the present invention when there is no pressure to force the polymer out of its insulating position, is a polyisobutylene having an average molecular weight as determined by the Staudinger viscosity method lying between about 3,000 and about 25,000 or 27,000. This material is a solid but very sticky substance, plastic but not readily melted, and is very resistant to oxidation, hydration, or attack by chemical agents, e. g., acids, alkalis, etc. When resistance to flow or displacement under pressure is desired, a polymer having a much higher molecular weight should be used, e. g., preferably at least 100,000.

Cellophane, as is well known, is a regenerated cellulose, produced by mercerizing cellulose such as cotton linters or specially purified wood fibre paper pulp. The cellulose pulp is preferably laid into thick sheets having a characteristic texture such like that of blotting paper, and the dried sheets are steeped in 15% sodium hydroxide solution, usually for a period of about 4 hours, whereafter the greater portion of the caustic solution is squeezed out, taking with it in solution most of the alpha cellulose. The steeped, moist sheets are then disintegrated, or crumbed, and aged for a period of time which is conveniently about 4 days. At the end of the aging period, the material is "tumbled" with a quantity of carbon disulfide, to form cellulose xanthate, which thereafter is dumped directly into a suitable quantity of caustic soda solution, preferably about 6% concentration, in which substantially the whole of the cellulose xanthate is soluble. The solution is then filtered and aged for another period, also usually about 4 days, and thereafter is coated, preferably upon a polished wheel. Immediately after the coating, the viscous coat on the wheel is coagulated by the application thereto of an aqueous solution containing approximately 25% of sodium sulfate and from 20% to 35% of sulfuric acid. The application of the acid solution abstracts the sodium from the viscose solution, frees the carbon disulfide and precipitates the dissolved cellulose as a regenerated cellulose or cellulose hydrate in the form of a relatively thin, transparent film. The film is desirably washed through several acid solutions, and then through water, to remove all of the acidic and alkaline bodies, to leave as pure a regenerated cellulose as possible.

For commercial uses, such as wrapping tissue, the cellulose film is frequently treated with glycerine or other flexibilizing agents. For the condenser use, however, this treatment is omitted and instead the film is dried as thoroughly as possible.

When so prepared, the cellophane film has a breakdown voltage or dielectric strength of approximately 80,000 volts per millimeter thickness (2,000 volts per mil thickness) and a dielectric constant of approximately 7. In comparison, the polyisobutylene, as above prepared, has a breakdown voltage of approximately 23,000 volts per millimeter thickness (600 volts per mil thickness) and a dielectric constant of approximately 2.3 to 2.5.

Both materials have very high resistivity values, that for polyisobutylene being greater than 10^{15} ohms per centimeter cube, and for cellophane being comparable or slightly lower.

The metal foil which is a portion of one of the embodiments of the present invention, may be of any of the usual forms of foil such as tin foil, either pure tin or alloyed with lead, or may be aluminum foil, either in pure form or in the various alloys used for this purpose. It is especially

important to have the metal foil free from pin holes, cracks or defects, although this precaution is not as important with the present invention as with waxed paper, because the polyisobutylene tends to fill small holes and so tends to prevent premature breakdown.

In the assembling of the condenser, for low voltage service, a single thickness of cellophane may be utilized. This is preferably coated upon both sides with the polyisobutylene. If material having a molecular weight near the bottom of the suggested range is used, it may be coated directly upon the cellophane, being sufficiently plastic to adhere directly in the form of a thin, homogeneous coating. Alternatively, if polymers of the higher range, or solid polymers of relatively low tackiness having molecular weights at the suggested high, desirable range are used, the material may conveniently be applied in solution in a volatile solvent, e. g., naphtha, liquefied gaseous hydrocarbons such as pentane, butane or even propane, carbon tetrachloride, etc. In either event the cellophane is preferably coated between rolls if the solid polymer is used, and thereafter is rolled directly upon a small mandrel with the interleaved foil which forms the plates of the condenser. It is, of course, desirable that two strips of coated cellophane be utilized, together with two strips of foil. The whole is desirably rolled into the cylindrical condenser 1 as shown in Fig. 1, and connecting tabs 2 and 3 to the respective strips of foil are provided.

The condenser then takes the form as shown in the enlarged view of Fig. 2 in which a sheet of cellophane 4 has coated thereon a layer of the polymer material 5 adherent thereto and similarly adherent to a layer of foil 6. Over the layer of foil 6 is a second layer of the polymer material 7 which is adherent to the foil, and also adherent to a second layer of cellophane 8. The farther side of the cellophane 8 is also coated with a layer of polymer 9 which is adherent both to the surface of the cellophane and to the foil 11. On the second side of the foil 11 is still another coating of polymer 12 which is adherent to the foil 11, and also adherent to the second turn of the first layer of cellophane 4 which in turn is covered with a continuation of the first coating of polymer 5, the first layer of foil 6, the second coating of polymer 7, and the second strip of cellophane 8 which is continued until the desired number of turns and area of foil electrodes have been rolled up.

The preferred embodiment, as above described, is a rolled or folded type of condenser. It is, however, possible to produce other forms, such as stacked rectangular pieces of foil and coated cellophane in alternation, although this form of condenser is of commercially much less consequence. Similarly, there are occasional uses in which a guard strip is required, and accordingly three or more strips of foil may be utilized in various adjacent relationships. In view of these facts, it will be obvious that many other embodiments of electrostatic condensers can be constructed utilizing foil, cellophane and the polymeric material.

It will be observed that during the rolling of the condenser, the plastic character of the polymer avoids the formation of air bubbles between the foil and dielectric, since under the rolling pressure all traces of air are effectively squeezed out of the condenser. This characteristic further improves the quality of the condenser, since it removes all danger of ionization of air within the body of the dielectric. Furthermore, the removal of all traces of air, even though it is readily accomplished with the present construction, is of much less consequence, since the polymeric material is exceedingly resistant to oxidation from either air or ozone, and small air bubbles remaining adjacent to the foil plates, even though they might be ionized, are without harm to the condenser, in sharp contrast and distinction to all other types of dielectric suitable for rolled condensers.

Thus the smooth layers of foil 6 and 11 are separated by three-layer dielectric bodies, each of the layers being homogeneous and free from focal points for voltage gradients and each being bounded by smooth surface planes at which only a minor change in slope of the voltage gradient curve occurs. Thus the lack of focal points for voltage gradient greatly increases the permissible voltage between electrode surfaces, and greatly increases the working voltage of the condenser for a given thickness of dielectric.

For high voltage condensers, where one thickness of cellophane and polymer is insufficient to withstand the applied voltage, a plurality of layers of cellophane may be utilized, the successive layers of cellophane being held together by thin interposed films of polymer substance. By this procedure any desired number of layers of cellophane may be utilized to build up a thickness of dielectric sufficient to withstand the desired voltage.

It is to be observed that cellulose is generally considered to be a high molecular weight carbonaceous material and is a branched, linear hydrocarbon chain compound containing a limited number of hydroxyl groups on the chain; likewise the polymer substance is a high molecular weight carbonaceous material consisting of a branched chain hydrocarbon but not containing any hydroxyl groups. (The polymer has a double bond between the last two carbons at one end of the chain although the whole molecule is relatively so large that this one double bond does not produce any substantial amount of chemical unsaturation in the compound.) This similarity in chemical structure results in dielectric constants for the two materials which are so nearly the same that the change in slope of the voltage gradient at the interface between the cellophane layer and the polymer layer is very small, thereby still further smoothing out the voltage gradient curve between plates, and increasing the permissible applied voltage.

Such a condenser with a single sheet of cellophane between sheets of foil is desirably produced with the cellophane thickness at a value of about 2½ to 3½ mils (thousandths of an inch) and the polyisobutylene layers are desirably each about ½ mil in thickness. Such dimensions produce a condenser which will give reliably continuous service on ordinary sine wave form A. C. at impressed R. M. S. voltages of 300 to 900, at a good safety margin, depending upon the presence, amplitude, and wave shape of transients, or other abnormal voltage surges.

When several layers of cellophane are interposed between the foil sheets the permissible voltage is increased more than proportionately, because of the better distribution of voltage gradients produced by the interposed film of polyisobutylene.

The operating voltages are raised above those ordinarily usable on paper condensers because of the smooth, even voltage gradients, and the absence of ionization phenomena, due to absence

of air bubbles, absence of voltage gradient concentration, and the high resistance of the materials to ionization, which is particularly characteristic of the polyisobutylene.

The above-described embodiments of the invention utilize preferably polyisobutylene having an average molecular weight lying between about 3,000 and about 15,000 to 27,000, since this range of molecular weights contains the most powerfully sticky and adhesive forms of the polymer. However, the molecular weight polymers above 27,000 are also usable, as mentioned above.

Another advantage of the combination is the sealing effect against moisture produced by the polyisobutylene for the benefit of the cellophane. The very high resistance of the polyisobutylene to water and moisture provides a very valuable protection to the electric strength and properties of the cellophane, which is not obtainable in other substances.

Alternatively, other plastic solid polymers are usable, e. g., polymers of 2 methyl butene-1, or of mixed olefins or even of lower olefins such as ethylene and propylene. It is also possible to use polymers made by polymerizing together iso-olefins and diolefins such as isobutylene and butadiene, or isobutylene and isoprene or chloroprene and also ethyl methyl ethylene (2 methyl butene-1) and butadiene or isoprene or chloroprene, under conditions to produce a plastic high molecular weight (i. e., above 1,000 or so) linear type. These copolymers contain usually less than 10% down to 1% or slightly under the diolefin, and show the interesting property of being chemically nearly saturated, having an iodine number less than 30 and usually less than 20 (in contrast to rubber, which has an iodine number of 340 to 360). The polymer is chemically reactive with elemental sulfur, when heated, especially in the presence of sulfurization compounds such as Tuads (tetra methyl thiuram disulfide), whereby they undergo a further condensation to a high tensile strength product. These polymers also are prepared by the use of a Friedel-Crafts type catalyst, preferably aluminum chloride, dissolved in ethyl or methyl chloride at temperatures from −50° C. down to −150° C. These polymers likewise may be applied to the cellophane sheet or to the foil, from solution in a suitable solvent such as naphtha, carbon tetrachloride, chloroform, etc., and the solution also desirably contains elemental sulfur in a proportion of approximately 5% of the polymer material, and also the above-mentioned Tuads in the proportion of ½% on the polymer material. This form of polymer adheres well to both the cellophane and the foil, and in addition has the property of being cured under heat and pressure to form a tough and powerfully adhesive layer between the foil and the regenerated cellulose sheet.

In this embodiment of the invention, the foil or the cellophane or both may be coated with a very thin layer of the polymer material including the sulfur and the sulfur compound, and the foil and cellulose sheet may then be rolled up together and thereafter cured by the application of heat and pressure at a temperature of 120° C. to 180° C. for a period of 15 to 60 minutes.

This embodiment of the invention produces an electrostatic condenser in which the successive layers of dielectric and foil are bound into a solid unitary whole by the cured copolymer material.

If desired, about 10 parts by weight of the polymer to be used may be mixed with or dissolved in 1 to 50 parts of one or more materials serving as hardeners, such as paraffin wax or petrolatum wax or various other natural or synthetic waxes sufficiently compatible with the polymer, or various resins, especially hard hydrocarbon resins of petroleum origin, e. g., those made from cracking coil tar or from vapor phase cracked petroleum hydrocarbon gases. Such hardeners are desirable addition agents when the polymer composition is to be used in making rigid condensers not subjected to severe jarring or vibration. For some purposes, as for low-cost, low-voltage condensers, bituminous materials, e. g., asphalt or tar preferably of petroleum origin, may be admixed with the polymer. In other cases, where flexibility is desired and ability to stand up under vibration and shock, rubber or rubber-like materials may be used instead of the hardeners mentioned above and the resulting composition may be vulcanized if desired; the rubber and polymer may be mixed either by milling or by dissolving the polymer alone or with paraffin wax for example, in a volatile solvent, e. g., naphtha, swelling comminuted rubber in the resultant solution, spreading on the cellophane or metal foil surface and evaporating the solvent, as described in my applications Serial No. 670,730, filed May 12, 1933; No. 704,747, filed December 30, 1933; No. 170,048, filed October 20, 1937; and No. 185,519, filed January 18, 1939, now abandoned, of which the present application is a continuation-in-part.

Not only is the invention of value in the construction of fixed electrostatic condensers, such as described hereinbefore, but owing to the exceptionally good adhesive and dielectric properties of applicant's polymer bonding agent in conjunction with metal foil and homogeneous cellulosic sheet material of the cellophane type, it is now possible for the first time to make satisfactory continuously variable electrostatic condensers by rolling together as shown in Fig. 10 of the drawing on a single mandrel **22** two separate strips of insulated metal foil **23** and **23'** which are concurrently unwound from suitable supply rolls **24** and **24'**. The mandrel **22** should have a gear **25** or other coupling means attached thereto so that it may be turned by a dial knob (not shown) likewise having a gear or other coupling means (not shown) attached thereto, said gears having a high ratio, such as at least 2:1 and preferably 4:1, 10:1 or more, so that one complete turn of the dial knob (which may be marked to cover the complete wave length scale desired) will cause the mandrel **22** to make the required number of revolutions to wind up the necessary amount of metal foil capacitance. There may also be provision (not shown), such as a spring tension or mechanical gears or pulleys to cause the rewinding of supply rolls **24** and **24'** when the dial knob and mandrel **22** are rotated in an unwinding direction. The insulation portion of the insulated metal foil strips **23** and **23'** may consist of any suitably flexible insulating material such as paper or cloth, preferably impregnated with a flexible dielectric such as paraffin wax or high molecular weight polymers or mixtures thereof containing resins of other suitable hardening agents, to prevent tackiness of the surface; however, the preferred modification of such strips **23** and **23'** is a laminated product in which metal foil is bonded to a thin homogeneous cellulosic material, such as cellophane, with a plastic polymer of the type described hereinabove, such as polyisobutylene having a molecular weight above

1,000, preferably above 5,000 and better still above 30,000, inasmuch as the higher polymer having a molecular weight above this latter figure shows substantially less cold flow properties. Modifying agents such as paraffin wax, petrolatum, etc. may be used, if desired, although in most cases will not be necessary or advisable.

In regard to such a continuously variable condenser, a special novel feature of this invention is the use of mandrel foil strips, such as 26 in Fig. 11, which are relatively wide at one end and relatively pointed at the other so that when two such strips are wound together on the mandrel 22, having the two pointed ends of said strips insulatingly attached to the mandrel, the result will be a continuously variable condenser which is relatively much more sensitive in the low capacity range than in the high capacity range, because a single turn of the mandrel at the pointed end of the metal foil strips will create only a small capacitance, whereas at the large end of the metal foil a single turn of the mandrel will add a relatively much greater electrostatic capacity. In using pointed or several shaped metal foil strips 26, it is of course possible and preferable for the sake of mechanical strength and to avoid tearing, to mount said metal foil strips on homogeneous cellulosic strips having uniform width throughout, or at least having sufficient width throughout its length, to withstand the mechanical strains imparted by the winding up on the mandrel 22 and the reverse winding on the supply rolls 24 and 24'.

The invention of a continuously variable condenser relatively more sensitive in the low capacity range may, if desired, be carried out without the use of any plastic adhesive or bonding agent, as shown in Fig. 12, by feeding onto the mandrel 22 plain metal foil strips from supply rolls 26 and 26' while simultaneously feeding onto the same mandrel strips of homogeneous cellulosic or other insulating material from supply rolls 27 and 27'. For such construction, the metal foil strips will have to have sufficient thickness and/or width to withstand per se all mechanical strains, and also, there should be sufficient friction drag or spring tension on all of the supply rolls 26 and 26', 27 and 27', that when the mandrel 22 is rotated in a winding-up direction all of the four strips will be wound tightly so as to provide regular increase in electrostatic capacity, because looseness and irregularity would cause undesirable sounds, such as squeals and static in radio apparatus using such condensers.

A further modification of the invention of a variable condenser relatively more sensitive in the low capacity range may be embodied in an air condenser of which a cross-section at right angles to the shaft is shown in Fig. 13. This illustrates the fixed plate 28 and the rotating plate 29, both being pointed in the low capacity end and wide at the high capacity end. The dotted outline 29' shows the position of the rotating plate 29 when turned to the high capacity position. The slight loss in total possible capacity, due to the plates 28 and 29 not overlapping 100% when in the high capacity position, is much more than compensated for in regard to the efficiency of the condenser as a whole by its increased sensitivity in the low capacity range.

The combination of foil, cellophane and polymer as above described is of very great advantage in the construction of electrostatic condensers. It is also of outstanding value in the construction of the device known as condenser type transformer bushings, which are particularly advantageous in the smaller sizes in connection with small high tension transformers. As is well known, the problem of carrying the wire leads to a transformer through the metal case is relatively difficult of solution, because of the concentration of potential gradient by the metal case of the transformer which leads to difficulties with ionization and other phenomena which tend toward an early breakdown of the insulating bushings carrying the leads to the condenser windings. This problem has been solved by the use of rolled dielectric bushings around the conductor leads with interposed layers of foil which serve to distribute the dielectric stresses. Large bushings are customarily made of foil paper and Bakelite. Small bushings have to the present been neglected. The present structure is particularly advantageous for the construction of small bushing suitable for lower power transformers such as instrument transformers, and particularly suitable for small power, high voltage transformers used for vacuum tube supply purposes.

The combination of foil, cellophane and polymer is also advantageous for many other uses such as the electrodes in ionization chambers of various types, and for many other purposes where a metal film carried upon an organic material film base is desirable.

The present embodiments of the invention utilize the regenerated cellulose material known as cellophane, but the invention equally includes other homogeneous cellulosic sheets, such as sheets or films of cellulose nitrate, cellulose acetate, such as that marketed under the trade name "Plastacale" and other cellulose esters and ethers. These materials do not ordinarily have as high a breakdown voltage as does the cellophane, but they are more easily prepared in thicker films, and are of somewhat higher tensile strength, for which reason they are preferable in some embodiments.

Thus the device of the invention provides a combination of a metal foil, a smooth transparent cellulosic sheet, and a polymeric material interposed between the foil and cellulosic sheet. The device of the invention is particularly advantageous for electrostatic condensers of various types and for electrodes of various types, but it obviously is useful for many other purposes.

For instance, thin sheets or foil of metals such as aluminum, tin, gold, copper, lead, steel, etc. are coated or combined with thin transparent sheets of regenerated cellulose, cellulose acetate, etc., by using isobutylene polymers (2,000–300,000 M. W.) as binder or adhesive. The laminated product is useful as wrapper, book binder, etc.

The cellophane is tough and prevents tearing of metal foil which may be exceedingly thin, even 1 5/10,000 inch, and also preserves luster and yet does not mask this luster or any design on the foil.

The polymer adhesive is not only colorless, flexible, and non-hardening, but is preferably transparent because it does not crystallize as does paraffin wax.

As an example of such a product, the following is given.

A 5% solution of a polymerized isobutylene (having an average molecular weight of about 5,000) in a benzene-naphtha mixture was spread in a thin layer on a thin sheet of colorless cello-

phane, and after the solvent had evaporated, this sheet of cellophane (about .001" thick) was laminated (sticky side down) onto a thin sheet of aluminum foil (about .0002" thick), the two sheets of material being rubbed together to insure exclusion of any air pockets and to make a laminated product which was well bonded together and perfectly smooth.

The polymer adhesive may be applied either to the metal foil or to the homogeneous cellulosic sheet. The good adhesiveness of the polymer, alone or in admixture with paraffin wax, petrolatum, or other suitable diluent or hardening agent, permits the use of a very thin film, and hence light weight of polymer. If desired, red, blue or other oil soluble dyes may be dissolved in the polymer or in a volatile solvent solution thereof so that the color effect will be visible through the transparent cellulosic material. Although it is preferable to apply the polymer in the form of a continuous film or layer, it may, if desired, be applied only in certain parts as required for sufficient bonding strength. Pigments or dyes may be applied in only certain locations, if desired to produce designs.

The metal foil itself may be in the shape of a design or may be cut out like a stencil and used for decorative or advertising purposes as by providing illumination behind the cut-out metal foil design, or for making special X-ray blanks of lead foil and cellophane with a round or other shaped hole in the foil to permit rays to pass through only in the desired location, the rest being blanked off for protection of parts not desired to be exposed to the X-ray.

The foil may be of any desired thickness and may have various degrees of polish, hardness, texture (mottled, embossed, etc.) or form (sheet, stencil, silhouette, etc.) and on the exposed side it may have printing or design or color as desired. Also, to increase its stability against oxidation, the metal foil may be given a preliminary oxide coating by any suitable method such as by exposing either one or both sides to an oxidizing fluid. The foil may also be coated with lacquer.

Laminated products made according to this feature of the invention may have any number of layers of metal foil and transparent cellulosic material; for instance, a product made of one layer of each may be used, preferably with the cellulosic sheet on the outside as a wrapper for packages of foods, etc., as a book covering material, as a flexible composite product impervious to gas, moisture, water and other liquids and to light; and it is also useful as a curtain or shade in stores, homes, autos, etc. These products may also be used as a heat-insulating material either in sheet form (to reflect heat rays from either one or both sides) or in a crumpled form (as it has been heretofore proposed to use aluminum foil). With a thin film of a transparent homogeneous cellulosic material bonded to the aluminum foil, there is the additional advantage that small surfaces of the crumpled metal foil are heat-insulated from each other by the cellulosic material and hence prevent heat conductivity, while at the same time giving heat reflection and good heat insulation by breaking up the air space into small pockets.

Similarly, products made with one sheet of metal foil in between two sheets of cellophane, or vice versa, one sheet of cellophane between two sheets of metal foil, may be used for similar purposes. If desired, a backing of paper, cloth, wood or other fibrous material may be provided, especially on the metal foil side of such laminated products; any suitable adhesive may be used for attaching this backing material and although the substantially saturated plastic polymers described hereinabove are preferred, other less expensive materials, such as bituminous products may sometimes be suitable. Also, non-penetrating adhesive compositions, such as described in my copending application 704,747, filed December 30, 1933, may be used.

In the various above described laminated products one or more edges thereof may be turned back and sealed down by a polymer or other suitable adhesive, or by a suitable volatile solvent solution of cellulosic material.

The polymer adhesive of this invention, i. e., polyisobutylene, etc., has such good adhesion properties that a much smaller amount of it can be used than would have to be used of the bituminous binder heretofore employed. Incidentally, polyisobutylene is practically unique in its adaptability for use in binding together two sheets of material (e. g., metal foil and cellophane), which are both relatively so impervious to vapor that they cannot be used commercially with a binder containing a solvent which must evaporate, as the latter cannot evaporate fast enough through the cellophane and cannot evaporate at all through the metal foil.

In preparing such laminated products, the regenerated cellulose or other transparent homogeneous cellulosic material may have a thickness ranging from about $\frac{1}{10}$ mil to 5 mils, preferably 1–3 mils (1 mil=$\frac{1}{1000}$"), when it is to be used for any flexible rolling or wrapping purpose, or the thickness may range on up to 100 mils or more where the intended use requires flexible but relatively stiff sheets. Although exceedingly thin metal foil may be used, as indicated previously, it is also possible to use substantially greater thicknesses thereof, e. g. $\frac{1}{64}$", $\frac{1}{32}$", or even relatively stiff sheets of metal of even greater thickness. The amount of the polymer adhesive to be used will, of course, vary to some extent, but ordinarily it will be within the approximate limits of 0.5 mg. to 10 mg./sq. cm. and will usually be within the limits of about 1–5 mg./sq. cm.

Instead of using metal foil, it may also be desirable at times to use finely divided metal such as aluminum powder and the like, in which case the material to be coated may first be given a thin coating of adhesive and then dusted or sprayed with the aluminum powder which may then be pressed onto the surface by any suitable means such as rolling, and the excess powder may be removed by blowing it away with an air blast or by brushing.

In the accompanying drawing, Figures 3–6 show several different modifications of laminated products prepared according to this invention, with part either cut away or turned back so as to reveal the construction of the product, showing the several laminated sheet materials and the adhesive. Fig. 3 shows a product made of one sheet of regenerated cellulose 4 bonded with polyisobutylene 5 to a sheet of metal foil 6; it also shows how one of the edges may be turned over for protection against tearing, as indicated at 14. Fig. 4 illustrates a laminated sheet material composed of a single sheet of regenerated cellulose 4 bonded on each side by polyisobutylene adhesive 5 to two sheets of metal foil 6 which may have either the same or different texture or mottled appearance or other

design, and may each be so thin that they would tear very easily if not supported by the intermediate sheet of regenerated cellulose. Fig. 5 shows more or less the reverse of the product shown in Fig. 4, namely, two sheets of regenerated cellulose 4 with an intermediate very thin sheet of metal foil 6 bonded on each side by polyisobutylene adhesive. In this case the smooth tough coating of regenerated cellulose protects the thin delicate film of metal foil from tearing. Fig. 6 is similar to Fig. 5 in having two outer sheets of regenerated cellulose which are transparent, and it may be either colorless or colored, bonded together with a layer of polyisobutylene adhesive containing finely divided aluminum powder 13 interspersed therein, either uniformly throughout the adhesive or only in certain locations to produce a certain designing or lettering or decorative effect.

Another feature of this invention which makes use of the unexpected advantages of the combination of unexcelled insulating properties and good adhesive characteristics, is a metal electrode insulated by transparent homogeneous cellulosic material, said element being bonded by a plastic substantially saturated linear hydrocarbon polymer. The electrical condenser first described above is one embodiment coming within the scope of this generic product. Another embodiment offering many unexpected advantages over products used heretofore is an electrical conductor such as a solid or stranded cable of copper or aluminum, insulated by regenerated cellulose in strip form, which may be applied either by the conventional spiral-wrapping method or by the strip-covering method; in either case a plastic polymer such as described hereinabove, for instance, polyisobutylene having a molecular weight above 1,000, preferably above 27,000 or 30,000, and preferably still above 100,000, is used as an insulating adhesive to bond the regenerated cellulose to the electric conductor and also to bond the several layers of the regenerated cellulose to each other with the exclusion of air pockets. Although regenerated cellulose is preferred here, other homogeneous cellulose materials may be used, such as ethyl cellulose, cellulose acetate, etc. Also, instead of the polyisobutylene, halogenated- or sulfur chloride- derivatives thereof may be used, and the texture of the bonding layer, i. e., the polymer, may be modified by non-volatile diluents, e. .g., paraffin wax, petrolatum, resins, oils, rubber, gutta percha, etc.

The polymer adhesive may either be applied as an integral layer, i. e., as a continuous film of the adhesive per se, or it may be applied in the form of a thin, preferably spirally wrapped, strip of fibrous material, such as paper or textile fabric impregnated with the polymer. If a film of adhesive per se is to be applied directly to the electrical conductor, it may be applied by various methods such as dipping, spraying, brushing, etc., using either a solution of the polymer in a material such as paraffin wax or gutta percha, and then letting the film cool, or by using a solution of the polymer in a volatile solvent, such as naphtha or liquefied gaseous hydrocarbons, and then letting the solvent evaporate. If desired, extrusion may also be used, especialy when the conductor in question is a stranded conductor, in order to fill any cracks, crevices, or other irregular surfaces in the conductor and completely surround it with a thin uniform film of the polymer adhesive. A strip of cellophane or other homogeneous cellulosic material may then be directly applied to the coated conductor either by spiral-wrapping or strip-covering. If desired, such application of adhesive and cellulosic insulating strip may be repeated a plurality of times until an insulation of the desired thickness has been applied. In such operations, the amount of polymer adhesive should not be more than necessary to give a thin continuous film of adhesive and should not be sufficient to permit any substantial displacement of the conductor with respect to the insulation due to any cold flow property of the adhesive.

Another method of carrying out this feature of the invention is to coat one side of a strip of cellophane or other homogeneous cellulosic material with a volatile solvent solution of polymer adhesive, e. g., polyisobutylene having a molecular weight, for instance, of about 80,000, then allow the solvent to evaporate completely (preferably hastening the continuous operation by vacuum oven drying) then wrap the strip spirally or by strip-covering around the electrical conductor to be insulated. Heat may be used to soften the coating during application to the conductor, especially if the halogenated derivatives of the polymer are used, e. g., a chlorinated polyisobutylene containing 20–50%, preferably 30–45% of chlorine. If desired, copolymeric adhesives may be used, especially thermoplastic copolymers such as those obtained by copolymerizing isobutylene and styrene at temperatures below −10° C. by an active halide catalyst such as boron fluoride.

For especially good insulating results, a combination of spiral-wrapping and strip-covering may be used, for instance, by applying alternate layers of the cellulosic insulation by these two methods, one after the other. Also, if desired, a solid high molecular weight polymer of ethylene or copolymers thereof with other olefins, such as isobutylene, or diolefins such as butadiene, made by polymerizing such materials under high pressures, e. g., 500 atmospheres or more, and at a temperature of about 100–250° or 300° C., preferably in the presence of a small amount of oxygen, may be used either alone, if sufficiently plastic and adhesive, or in admixture with some of the more plastic and adhesive polymers, e. g., polyisobutylene, described hereinabove.

The polymer adhesive described above may also be mixed with rubber, if desired, and then the resultant mixture of polymer and rubber may be applied to the electrical conductor either by extrusion, or if preferred in the form of a self-sustaining strip, it may be applied by either the spiral-wrapping or strip-covering method, and then the outer insulating layer of homogeneous cellulosic material applied. For such purpose the rubber and polymer, alone or together with any consistency modifiers, such as waxes, resins, oils, etc. may be mixed together on steel rolls such as are used in the ordinary rubber mill, or they may be kneaded together in a suitable apparatus, such as the Banbury mixer or the composition may be built up by laminating together one or more layers of the rubber and polymer. However, the preferred method of compounding the polymer and rubber is the method described in my copending application 704,747, which discloses dissolving, for example, 1–10% of an isobutylene polymer in gasoline and swelling rubber (in the form of finely divided particles or small pieces, preferably having a thickness not greater than about ⅛" or 2 or 3 mm.) in the resultant solution. The rubber is allowed to swell until the

particles of rubber become flabby and jelly-like in texture but not sufficiently that they break down and lose their inherent colloidal structure. When the swelling has proceeded to the desired extent, any residual unabsorbed solution of the polymer and gasoline is preferably removed by decantation, filtration, and/or washing with fresh gasoline, and the residual swollen jelly-like particles of rubber aggregate may be agitated sufficiently to produce a substantially homogeneous and smooth viscous composition. A slight mechanical comminution in any suitable manner (as by forcing the composition through a 1/16" wire mesh screen) may be used if desired. When the resulting viscous composition is allowed to evaporate, the residual dry rubber-polymer composition is found to be a homogeneous mixture of the high molecular weight saturated hydrocarbon polymer intimately and uniformly or homogeneously dispersed within the colloidal particles or cells of the rubber. Instead of using gasoline, naphtha or other suitable volatile liquids may be used which act as solvents for the polymer and/or wax and similar materials, but only as swelling agents for the rubber.

The electric conductor to be insulated may consist of wires or cables of either the solid, stranded or braided types or may be metal in flat or curved sheet forms such as used in the construction of condensers and the like. The conductors may be composed of the usual metals such as copper, aluminum, silver or alloys thereof, or they may consist of a thin deposit or coating of a metal having a high electric conductivity on a metal having a lower electric conductivity such as a copper wire electroplated with copper, or iron or steel wire with an outer layer of extruded aluminum. Also, if desired, the electric conductor may have already been given an insulated coating of enamel or particularly in the case of aluminum it may have been subjected to an oxidizing treatment in order to coat the aluminum conductor with a film of aluminum oxide which serves as insulation.

In the accompanying drawing Fig. 7 illustrates an electrical conductor **15** coated with a polyisobutylene adhesive layer **16**, then spirally wrapped with regenerated cellulose in strip form **17**, an overlying insulating layer of vulcanized rubber **18**, and a final outer covering of braid **19**.

Another special feature of this invention is the use of a thin continuous film of polyisobutylene or other plastic polymer of the type described, as an immediate protective covering on an oxide-coated aluminum conductor or cable, especially when such cable is to have an overlying insulating layer of rubber or homogeneous cellulosic material such as cellophane. In this modification of the invention, the film of polymer (alone or together with consistency modifiers, such as paraffin wax, resin, gutta percha, etc.), due to its special resistance to oxidation, protects the oxide coating against acidic or other chemical influences tending to remove part of the oxygen or otherwise impair the oxide coating, and it thereby also simultaneously prevents the oxygen from said oxide coating from chemically combining with the rubber or other overlying insulating material and thereby prevents deterioration of the latter. The polymer thus serves a doubly valuable purpose. For such application, it is preferable to use polymers of relatively high molecular weight such as polyisobutylene of more than 30,000 molecular weight and preferably 100,000 or more. It is especially desirable for this purpose to use polymers which have been subjected to selective extraction or selective precipitation in order to obtain fractions which are substantially free from any polymers having a molecular weight below the desired minimum, because polymers having a molecular weight as low as 5,000 or 10,000 tend to have a solvent action on the rubber and eventually become miscible therewith so that in time, some of the rubber molecules, even though microscopically admixed with the saturated polymer, come directly in contact with the oxide coating on the metal conductor and tend to chemically combine with some of the oxygen in said coating, thereby impairing its electrical insulation value.

In the accompanying drawing, Fig. 8 illustrates an electrical conductor **15**, made for instance of aluminum, having an oxide coating **20** thereon, and a protective coating **16** comprising polyisobutylene, e. g., having an average molecular weight of about 150,000, alone or together with a thermoplastic material such as paraffin wax, and a thick insulating layer of vulcanized rubber **18** to protect the conductor from mechanical shock or abrasion, and finally the conventional outer covering of braid **19**. If instead of rubber, cellophane is used in true form, it may be applied by the spiral-wrapping method or strip-covering method by any one of the several procedures outlined above.

The above discussed concept of using a film of polyisobutylene or equivalent polymer to protect an oxide-coated aluminum conductor may be expressed more generically as the protection of any oxide-coated metal surface by a thin continuous film comprising a plastic substantially saturated linear hydrocarbon polymer. Another specific modification of this broad idea is the protection of oxide-coated sheet iron with a thin continuous film comprising high molecular weight polyisobutylene having a molecular weight of at least 1,000, preferably 5,000 or more, alone or together with paraffin wax or other substantially non-oxidizable texture modifiers or hardeners. For instance, it is recognized that sheet iron and other ferrous products, if given an oxide coating by treatment with oxygen or chemical oxidizing agent, especially under elevated temperatures, will help to protect the metal and make it more durable under the severe conditions under which it is to be actually used, but oftentimes such oxide-coated iron products are subjected to acid fumes or other chemical influences which tend to impair the oxide coating during storage or transportation and under such conditions a thin film of the saturated polymers of this invention serve to protect the oxide coating on the iron until the latter is actually ready to be used in its proper industrial application. This protective film of polyisobutylene or other polymer may be readily removed from the oxide-coated metal surface by a light washing or sponging with a solvent such as naphtha, if desired, or may be permitted to remain on the oxide coated metal surface during use if not actually incompatible under the conditions in which the metal is used.

In Fig. 9 in the accompanying drawing, a piece of sheet iron **21** having an oxide-coating **20** and an overlying protective film **16** comprising polyisobutylene, is shown.

While there are above disclosed but a limited number of the embodiments of the structure of the invention, it is possible to produce still other embodiments without departing from the inventive concept herein disclosed, and it is therefore de-

sired that only such limitations be imposed upon the appended claims as are stated therein or required by the prior art.

The invention claimed is:

1. A copper conductor and a plurality of layers of spiral-wrapped regenerated cellulose sheet insulating material, bonded thereto by a thin film comprising polyisobutylene serving to bond the cellulose sheet to the surface of the conductor and to bond the plurality of layers of the cellulose together.

2. In combination an oxide-coated aluminum conductor, an insulating layer of vulcanized rubber, and between said oxide-coated aluminum conductor and said rubber insulation, a thin, continuous film of a plastic substantially saturated linear hydrocarbon polymer and a continuous cellulosic film bonded to said conductor by said polymer, said polymer having a molecular weight sufficiently high that said polymer has substantially no solvent effect on said rubber.

3. Conductor according to claim 2 in which the polymer is a polyisobutylene having a molecular weight of at least 30,000.

4. A copper conductor and a plurality of layers of spiral-wrapped regenerated cellulose sheet insulating material, bonded thereto by a thin film comprising a copolymer of polyisobutylene and a polyolefin, combined with sulfur, serving to bond the cellulose sheet to the surface of the conductor and to bond the plurality of layers of the cellulose together.

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