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SELF-SEALING INSULATING MATERIAL OF
HIGH DIELECTRIC STRENGTH

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This invention relates to materials of high dielectric strength suitable for insulating electrical conductors and the like. It relates particularly to an improved composition in tape, sheet or mass form which has the property of sealing itself into a substantially homogeneous body of high electrical insulating value when applied to a conductor or other article.

The flexible or resilient insulating materials provided heretofore generally have been rubber-base materials and have long been known to have certain undesirable characteristics. While such rubber-base materials have excellent insulating properties, they nevertheless deteriorate quickly under some conditions of use. Thus, many of these materials become hard and crack when subjected to relatively high or relatively low temperatures. All of them deteriorate quickly in the presence of ozone. For this reason, great difficulty has been encountered in the past in properly insulating conductors which are used for or near high voltage lines or other equipment where corona discharge, arcing or other ozone generating conditions are present. This is equally true of the rubber insulating tapes which are widely used to form a rubbery sheath around a conductor or a connection. Such rubber tapes are convenient to use, but inasmuch as they consist principally of unvulcanized rubber and an accelerator, they have the disadvantage of being self-vulcanizing so that their shelf life is relatively short. Also, these tapes are sticky so that the tape tends to stick together, often times tearing and making its application to the wire or other article being insulated a rather difficult and messy operation.

The present invention overcomes the disadvantages of these prior flexible insulating materials by providing a composition which is unaffected over very long periods of time by high concentrations of ozone. It is stable and thus has a very long shelf life. It is not especially tacky and thus does not tend to stick together very strongly. In addition, it has dielectric properties equal to or better than the rubber tapes and insulating materials produced heretofore. Moreover, my new composition, when provided in tape or sheet form, has the highly desirable property of being self-sealing, that is, when the material is stretched and then wrapped around a conductor

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or other article, it has a "creep" or "memory" such that it contracts and flows together to form a homogeneous body which cannot be delaminated after setting for a relatively short period of time. This material, upon setting, does not show laminations corresponding to the convolutions of the material. Thus, if the tape is wound around a conductor in the usual way, a continuous sheath is provided which does not have a junction line or zone between the overlapping portions of the material.

The properties of the material can be varied substantially, depending upon its intended use. Thus, the material may be made very pliable and soft for use under low temperature conditions wherein ordinary insulation becomes brittle and shatters. On the other hand, its properties can be modified so that it is resistant to high temperatures and it will adhere to and remain on the conductor without any tendency to flow away or to drop from the conductor. Compositions of optimum characteristics which are capable of withstanding a very great range of temperatures can be produced and thereby serve effectively from below zero to temperatures well above 100° F.

A particularly novel characteristic of my new composition is that it discloses visually the optimum degree of stretch to be attained before applying the material to the article to be insulated. In the preferred type of composition, the tape changes color when it is stretched sufficiently to produce the memory or creep effect which renders the tape self-sealing.

Generally, tapes of the type embodying the present invention and having the most desirable properties for commercial use consist of a compatible mixture of at least four components. These components are polyethylene of a relatively high molecular weight, polymerized isobutylene of an intermediate molecular weight or a mixture of isobutylene polymers some of which are of high molecular weight and others of relatively low molecular weight, butyl rubber, i. e., a sulfur-vulcanizable rubbery copolymer of isobutylene and a diolefin in approximately the proportions of 98:2, and a resinous tackifier.

The polyethylene component may include a small proportion of an anti-oxidant such as an aromatic amine. A typical anti-oxidant may

consist of 35% diphenyl-paraphenylene diamine and 65% phenyl-alpha-naphthyl-amine.

The property of resistance to deterioration by the action of ozone is believed to be largely due to the fact that this composition contains very little unsaturated material, the only unsaturated compound present being isoprene. Thus, there are few linkages available for coupling with oxygen which would cause a change in or breaking down of the plastic and insulating properties of the composition.

The functions of the various components of my new insulating composition are believed to be as follows: The polyethylene is in the nature of an inert filler having good insulating properties. It also serves to indicate the proper degree of stretch of the finished tape or material. Polyethylene is a good insulator but it is semi-rigid when of the molecular weight preferred in the composition and it has relatively low physical strength.

Butyl rubber is highly compatible with polyethylene and has the function of reducing the rigidity of the polyethylene and providing increased stretch in the material.

Polymerized isobutylene increases the tear strength of the material and also appears to increase the resistance of the composition to oxidation, especially oxidation by ozone. It also, when used in the proper molecular weight, reduces the rubberiness produced by the butyl rubber.

The resinous tackifiers referred to above produce a plasticizing action and cause the material to flow under pressure, thereby making the material bond together. The resinous tackifier may be any of a large group of synthetic or natural resins or derivatives thereof which are recognized for their ability to impart tackiness to resinous products. Among those resinous tackifiers that have been used in my new composition are cumarone-indene resins, alpha methyl styrenes, phenol-formaldehyde resins, alkyd-type resins, terpene resins, wood rosins, rosin esters, such as ester gums, hydrogenated diterpenes, pentaerythritol abietates, hydroabietyl alcohol, dehydroabietic acid, zinc resinates, gutta percha resin derivatives and many others.

The best results are obtained with abietic acid derivatives of the types referred to above. The preferred resinous tackifier is dehydroabietic acid which has a specific optical rotation in benzene of 49.5, an acid number of 158 and a saponification number of 164.

The action of the polyethylene in providing an indication of stretch is not clearly understood but it is believed to be a function of the long chain relation between the various components whereby stretching of the composition forces some of the polyethylene out of the mixture or solution and thereby causes the composition to change from a greenish to a whitish color.

The proportions of the various components of my new composition can be varied substantially while at the same time permitting the production of an acceptable insulating material. The polyethylene, being in the nature of an inert filler, can be varied within a relatively wide range and might even be omitted from the composition if it were not for two drawbacks resulting from such omission. The first of these is that the resulting composition would not have any means for indicating the optimum stretch of the material and thus it would be difficult to determine when the tape has been stretched enough to put it in condi-

tion for proper sealing. The polyethylene also acts to restrict the creep of the other materials and in this way controls the creep of the composition.

The other materials cooperate with the polyethylene to reduce the effect of thermal shock on the polyethylene component. Polyethylene, alone, if heated and then chilled abruptly, would shatter.

The butyl rubber component has the effect of imparting rubberiness to the composition and also tends to provide physical strength which is lacking in the polyethylene. Thus, if butyl rubber were omitted from the composition, the resulting composition does not stretch at all, and even in the presence of 5% of butyl rubber, the material is not very resilient and is short, that is, has a tendency to break upon stretching.

When the proportion of butyl rubber is increased to above 25%, the resulting material is quite weak and is difficult to process and handle.

Usually, the composition contains a mixture of low molecular weight polymerized isobutylene and a higher molecular weight polymerized isobutylene. Here again, the proportions of these components are susceptible to considerable variation. When the low molecular weight isobutylene polymer is absent, the material lacks tackiness and is short and breaks easily. This condition obtains even when as much as 4% of the low molecular weight isobutylene polymer is present.

When more than 25% isobutylene polymer (low molecular weight) is present, the composition becomes very tacky and flows when heated, so much so, that a composition containing 30% of the lower molecular weight polymer cannot be processed on a calender at all for it sticks to the rolls and cannot be removed.

The higher molecular weight isobutylene polymer has a tendency to reduce the stretch of the composition but it improves the resiliency of the composition. In the absence of this material, the composition is quite weak and short, i. e., it breaks easily, and even when as much as 5% of this material is present, this condition still exists to some degree. When as much as 40% of the higher molecular weight isobutylene polymer is present, the material becomes very tough, does not stretch easily and has a hard, horny appearance. The combined amounts of the isobutylene polymers can be varied, accordingly, between about 9% and 65% of the composition.

The isobutylene polymers are commercially available products identified by the molecular weights as given above. The molecular weights of these polymers are determined by the Staudinar method of determining intrinsic viscosity and viscosity average molecular weight as disclosed in the Standard Inspection Laboratory Circular No. 320.01 dated February 28, 1944.

The tackifier produces the tackiness and sealing effect which characterizes the new composition. If the tackifier is omitted, the composition stretches but does not seal readily. When about 1% of the tackifier is added, the extensibility or stretch of the material is increased substantially and the tackiness is increased sufficiently to provide some sealing action. The maximum amount of the tackifier is limited to about 20% inasmuch as, above this proportion, the tackifier renders the material very sticky and short so that it is difficult to process. When as much as 25% of the tackifier is present, the material becomes so tacky or sticky that it cannot be processed on a calender roll to reduce it to sheet or tape form.

From the preceding description, it will be apparent that the proportions of the several components may be varied substantially while still providing a self-sealing insulating composition which is satisfactory for use under many conditions. However, for commercial use and production, the best results are obtained when the composition contains components in about the following proportions:

	Per cent
Polyethylene, about 20,000 molecular weight -----	50
Butyl rubber -----	13½
Polymerized isobutylene, 100,000 to 115,000 molecular weight-----	22½
Polymerized isobutylene, 10,000 to 20,000 molecular weight-----	10
Resinous tackifier -----	4

This composition stretches readily but is quite tough so that it breaks only when subjected to very strong stretching forces, it has a slightly tacky but not sticky surface and is of a faintly greenish color when unstretched, changing to a whitish color when stretched sufficiently to render its creep effective for sealing action. The material recovers slowly, unlike rubber, but nevertheless will return substantially to its unstretched condition when relieved of tension. It is the pressure exerted by the creep of the material which apparently causes the material to flow together and thereby bond or cohere so completely as to form a substantially homogeneous body.

The composition is susceptible to further modification. Thus, for example, a single isobutylene polymer can be used instead of the isobutylene polymers described in the specific example and the examples given above. The average unit molecular weight of this single polymer should be about the same as the average of the combined unit molecular weights of the two isobutylene polymers. Thus, for example, an isobutylene polymer having a molecular weight of about 60,000 to 75,000 can be substituted in an equal amount by weight for the combined amounts of the two isobutylene polymers referred to in the specific example. The ultimate result is about the same for it appears that the degree of polymerization of this material controls its strength imparting and resilient properties.

From the preceding description of typical compositions embodying the present invention, it will be apparent that I have provided a composition which has the stability under variations of temperature and in the presence of ozone which is lacking from the prior rubber tapes and rubbery insulating materials. My new composition is useful under conditions where the prior rubbery insulating materials are not satisfactory. Moreover, it has the additional property of improved appearance due to its color. It provides a stronger bond between the various laminations of the material and it has a far greater shelf life than prior insulating tapes and the like.

As indicated above, the material is susceptible to considerable modification in the proportions of its several components, and, therefore, the example given above should be considered as illustrative of the invention and not as limiting the scope of the following claims.

I claim:

1. An insulating composition of high dielectric strength which upon being stretched and disposed in overlapping laminations flows together into a homogeneous body, consisting essentially

of a normally semi-rigid polyethylene having electrical insulating properties, the amount of polyethylene being up to 50%, between about 5% and 20% of a sulfur-vulcanizable rubbery copolymer of isobutylene and a diolefin in approximately the proportions of 98:2, between about 9% and 65% of at least one polyisobutylene having an average unit molecular weight between about 60,000 and 75,000 and between about 1% and 20% of a resinous tackifier for imparting flow and cohesiveness to said composition to cause said laminations to flow together.

2. An insulating composition of high dielectric strength which upon being stretched and disposed in overlapping laminations flows together into a homogeneous body consisting essentially of polyethylene having a molecular weight of about 20,000, the amount of polyethylene being up to 50%, between about 5% and 20% of a sulfur-vulcanizable rubbery copolymer of isobutylene and a diolefin in approximately the proportions of 98:2, between about 9% and 65% of polyisobutylene polymer having an average molecular weight between about 60,000 and 75,000 and between about 1% and 20% of a resinous tackifier of the class consisting of rosin, rosin esters, dehydroabietic acid, zinc resinate and hydroabietyl alcohol.

3. An insulating composition of high dielectric strength which upon being stretched and disposed in overlapping laminations flows together into a homogeneous body consisting essentially of about 50% polyethylene having a molecular weight of about 20,000, about 13.5% of a sulfur-vulcanizable rubbery copolymer of isobutylene and a diolefin in approximately the proportions of 98:2, about 22.5% of a polyisobutylene having a molecular weight between about 100,000 and 115,000, about 10% of a polyisobutylene having a molecular weight between about 10,000 and 20,000 and about 4% of a resinous tackifier for imparting flow and cohesiveness to said composition to cause said laminations to flow together, said composition having the property of changing color upon stretching to indicate the proper degree of stretch before laminating.

4. An insulating tape having high dielectric strength, said tape being stretchable, resilient, changeable in color upon stretching and flowable into a homogeneous mass upon stretching and winding into a plurality of overlying convolutions, said tape consisting essentially of a normally semi-rigid polyethylene, the amount of polyethylene being up to 50%, about 5% to 25% of a sulfur-vulcanizable rubbery copolymer of isobutylene and a diolefin in approximately the proportions of 98:2 for imparting resiliency to said polyethylene, about 9% to 65% of at least one polyisobutylene having an average unit molecular weight between about 60,000 and 75,000 for controlling the resiliency of the composition, and about 1% to 20% of a resinous tackifier for increasing the tackiness and flowability of the composition to cause the laminations to bond and flow together.

5. An insulating composition of high dielectric strength in sheet form which upon being stretched and disposed in overlapping laminations flows together into a homogeneous body, consisting essentially of about 50% polyethylene having a molecular weight of about 20,000 about 13.5% of a sulfur-vulcanizable rubbery copolymer of isobutylene and a diolefin in approximately the proportions of 98:2, about 22.5% of a polyisobutylene having a molecular weight between

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about 100,000 and 115,000, about 10% of polyisobutylene having a molecular weight between about 10,000 and 20,000 and about 4% of a resinous tackifier of the class consisting of rosin, rosin esters, dehydroabietic acid, zinc resinate, and hydroabietyl alcohol for imparting flow and cohesiveness to said composition to cause said laminations to flow together, said composition having the property of changing color upon stretching to indicate the proper degree of stretch before laminating.

6. An insulating composition of high dielectric strength, in tape form which upon being stretched and disposed in overlapping laminations flows together into a homogeneous body, consisting essentially of about 50% of a normally semi-rigid polyethylene having electrical insulating properties, about 13.5% of a sulfur-vulcanizable rubbery copolymer of isobutylene and a diolefin in approximately the proportions of 98:2, about 22.5% of polyisobutylene having a molecular weight between about 100,000 and 115,000, about 10% of

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polyisobutylene having a molecular weight between about 10,000 and 20,000, and about 4% of dehydroabietic acid, said composition having the property of changing color upon stretching to indicate the proper degree of stretch before laminating.

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