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Yasui et al.

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[45] **Apr. 17, 1973**

[54] **ELECTRICAL APPARATUS WITH THERMALLY STABILIZED CELLULOSE INSULATION**

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[73] Assignee: **Hitachi, Ltd.,** Tokyo, Japan

[22] Filed: **Apr. 5, 1972**

[21] Appl. No.: **241,114**

Related U.S. Application Data

[63] Continuation of Ser. No. 106,929, Jan. 15, 1971, abandoned, which is a continuation-in-part of Ser. No. 786,089, Dec. 23, 1968, abandoned.

[30] **Foreign Application Priority Data**

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July 5, 1968 Japan 43/46926
July 5, 1968 Japan 43/46927

[52] **U.S. Cl.**..................**174/17 LF,** 162/138, 252/64, 336/58, 174/25 R

[51] **Int. Cl.** **H01b 7/02**

[58] **Field of Search** ....174/17 R, 17 LF, 174/25 R; 162/138; 336/58; 252/64

[56] **References Cited**

UNITED STATES PATENTS

3,224,902 12/1965 Sadler....................162/138 X

Primary Examiner—E. A. Goldberg
Attorney—Paul M. Craig, Jr. et al.

[57] **ABSTRACT**

An electrical apparatus containing an insulating oil which is in contact with a cellulosic insulating material improved in heat resistance and stability by incorporation of a urethane compound containing at least one urethane bond and having a molecular weight of 75 to 1000.

8 Claims, 7 Drawing Figures

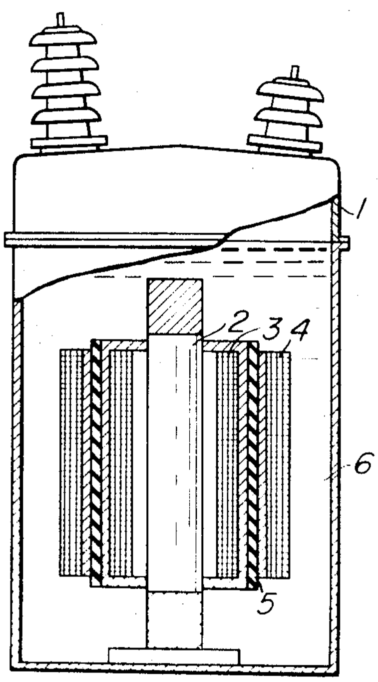


FIG. 1

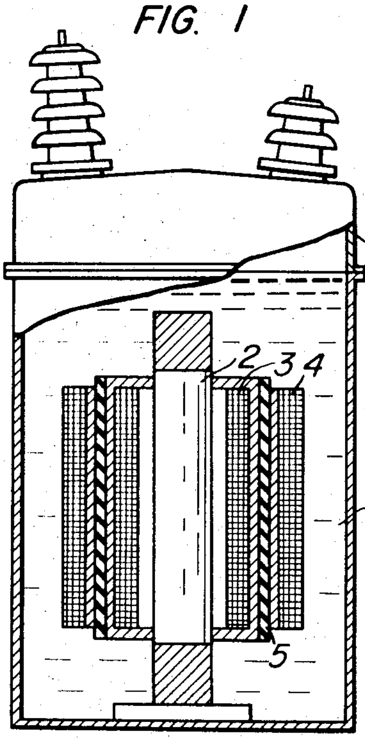


FIG. 2

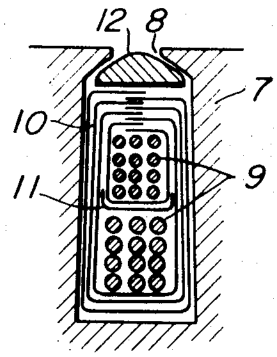


FIG. 3

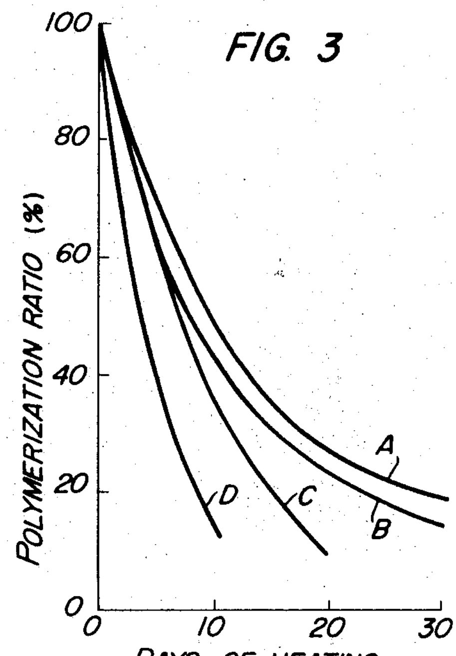


FIG. 5

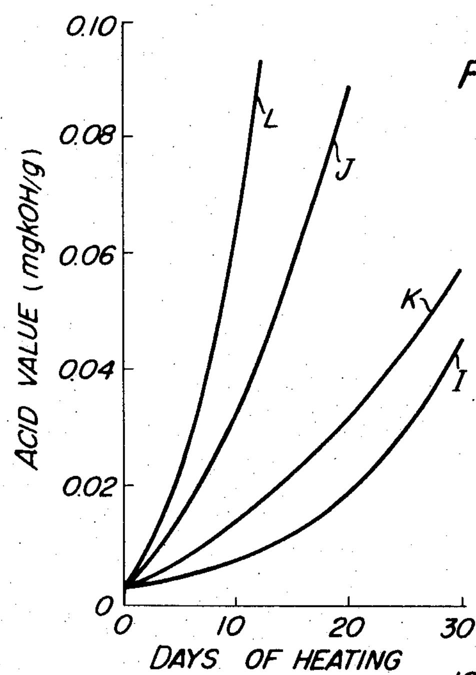

0.10
0.08
0.06
0.04
0.02
0
ACID VALUE (mgKOH/g)
L
J
K
I
0
10
20
30
DAYS OF HEATING

FIG. 4

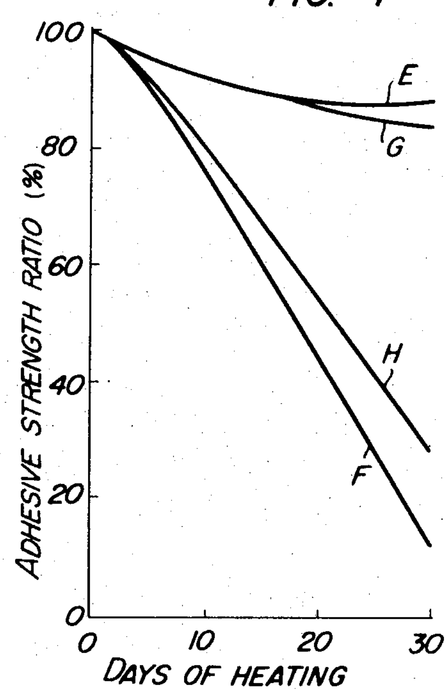

100
80
60
40
20
0
ADHESIVE STRENGTH RATIO (%)
E
G
H
F
0
10
20
30
DAYS OF HEATING

FIG. 6

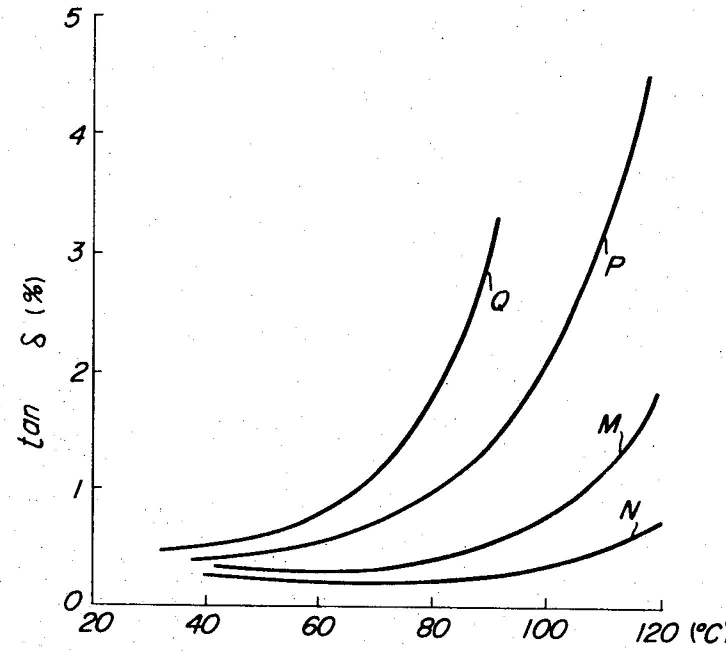
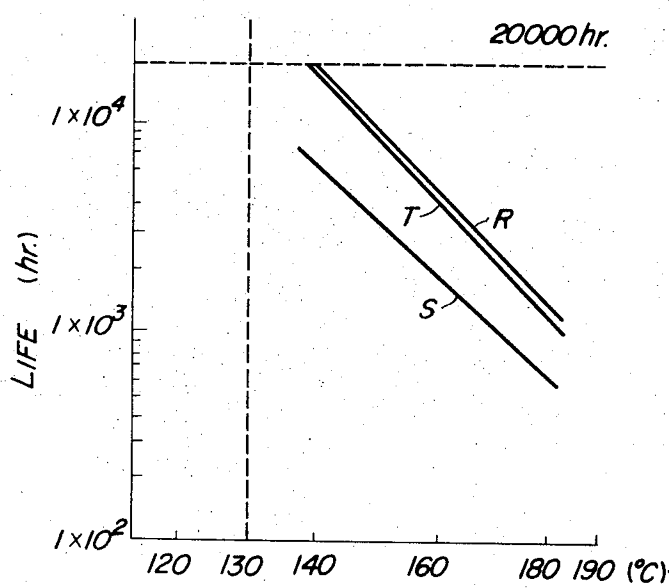

tan δ (%)
0
1
2
3
4
5
20
40
60
80
100
120 (°C)
Q
P
M
N

FIG. 7


20000 hr.
LIFE (hr.)
1×10^4
1×10^3
1×10^2
120
130
140
160
180
190 (°C)
T
R
S

ELECTRICAL APPARATUS WITH THERMALLY STABILIZED CELLULOSE INSULATION

This is a continuation of our application, Ser. No. 106,929 filed Jan. 15, 1971 now abandoned which in turn was a continuation of our application, Ser. No. 786,089 filed Dec. 23, 1968 and now abandoned.

In the case of transformers, for example, there have already been developed many magnet wires and insulating varnishes to be used in combination with insulating papers which are so excellent in heat stability as to be of the class E or class B (according to AIEE STANDARD NO. 510). In power transformers of the nitrogen gas-sealed type or completely sealed type, little problem takes place even when an insulating oil is used at a considerably high temperature. Further, when there is used an oxidation-resistant oil employed at present, it is possible to use the oil at a high temperature even in pole or distribution transformers of the ordinary sealed structure. However, in the case of the conventional oil-filled transformers, the temperature increase of the coils is restricted to below 55°C. More particularly, when the external air temperature is 40°C, the average coil temperature is restricted to 95°C and the hottest point is restricted to below 105°C. This is because the heat resistance and stability of cellulosic insulating materials employed such as insulating papers is of the class A even in an oil-filled state, and the maximum temperature thereof is limited to below said temperature.

From such viewpoints as mentioned above, there have been proposed several processes for improving the heat resistance of insulating papers, which are most low in heat resistance and stability among insulating materials for transformers, and thus a so-called 65°C transformer, in which the maximum service temperature increase of the transformer coil has been elevated by 10°C, has already been put into practical use.

As mentioned above, the maximum service temperature of a transformer can be elevated to a considerably high temperature by improving the heat resistance of the cellulosic insulating materials employed therein. This is not particularly limited to the case of transformers but is the same with the case of other electrical equipment.

Processes for improving the heat resistance and stability of cellulosic insulating materials may be roughly divided into two. One is a so-called partially chemical modification process, in which the hydroxyl group of cellulose is substituted with another group by chemical modification, while the other is a chemical treatment process, in which a heat resistance-improving additive is incorporated into a cellulosic insulating material. Cyanoethylated papers, acetylated papers and the like are insulating papers prepared according to the former process, i.e. the partially chemical modification process, and amine compound-treated papers are those prepared according to the latter process, i.e. the chemical treatment process. A comparison between the two processes shows that the latter process is substantially advantageous over the former process in that the costs for preparation are low and the properties inherent to cellulosic insulating materials are not injured. For the above reasons, the actual circumstances are such that the chemical treatment process is deemed to be of particular importance and is studied considerably preponderantly.

One of the objects of the invention is to provide an improved electrical insulating means comprising cellulosic insulating materials and a urethane compound provided in such a manner that the urethane compound has such an interaction with the cellulosic material as to improve the stability of the cellulosic material with respect to heat.

Another object of the present invention is to provide cellulosic insulating materials excellent in heat resistance and stability.

Another object is to provide cellulosic insulating materials which have no detrimental effect, both physically and chemically, on other coexisting insulating materials, particularly insulating oils, insulating enamel wires and insulating mold resins, and which do not promote the degradation in properties of said other insulating materials.

A further object is to provide cellulosic insulating materials which can be most successfully applied to electrical equipments when used in combination with other insulating materials, particularly insulating oils.

Another object is to provide cellulosic insulating materials which are useful for application to dry type electrical equipment.

A further object is to provide cellulosic insulating materials which can be easily prepared on a commercial scale with economical advantages.

A still further object is to provide cellulosic insulating materials which are useful as insulating materials for various electrical equipment over a considerably wide scope including such large size (large capacity) electrical equipment as oil-filled transformers, oil-filled condensers, oil-filled cables and generators, and such small size (small capacity) electrical equipment as small size transformers and general purpose motors.

Other objects will be apparent from the following explanation, taken in conjunction with the accompanying drawings.

The gist of the present invention resides in that a urethane compound having a molecular weight of 75 – 1000 is present together with a cellulosic insulating material.

In the accompanying drawings:

FIG. 1 is a partial longitudinal section of a distribution transformer which shows an application example of the present cellulosic insulating material.

FIG. 2 is a rough cross-sectional sketch of the stator slot portion of a revolving apparatus which shows another application example of the present cellulosic insulating material.

FIG. 3 is a graph showing the relationship between polymerization ratio

(average molecular weight of cellulosic materials after heating/original average molecular weight of cellulosic materials × 100) (%)

and heating time (days) of samples of insulating papers.

FIG. 4 is a graph showing the relationship between adhesive strength retention of enamel film (%) and heating time (days) of magnet wire-insulating films subjected to glass tube tests (heat degradation test together with samples of insulating papers).

FIG. 5 is a graph showing the relationship between acid values and heating days of insulating oils subjected to the glass tube tests.

FIG. 6 is a graph showing the relationship between dielectric dissipation factor (tan δ) and temperatures of samples of insulating papers.

FIG. 7 is a graph showing the relationship between lives and temperatures of samples of insulating papers, which indicates the results of moterette test (according to AIEE STANDARD NO. 510) of said samples.

This invention depends on a discovery of the fact that a heat degradation of the cellulosic insulating material, such as kraft paper, Manila paper, craped papers thereof, and the like may be considerably restrained by employment of a urethane compound, which is illustrated in the following description. According to the invention, the cellulosic insulating material is used in such a manner that the cellulosic material is affected by the urethane compound, that is, the urethane compound may catch or neutralize certain acid substances which occur by the heat degradation of the cellulosic material so that the heat degradation thereof may be prevented and moderated sufficiently.

In an oil-filled type transformer, the cellulosic insulating materials surrounding the conductors are used by incorporation with the urethane compound therein, and also used by immersing the urethane compound enveloped in a permeable bag in the insulating oil.

However, in the following detailed description, the insulating means according to the invention is described in the cellulosic insulating materials containing the urethane compound as typical examples.

In order to attain all the objects of the present invention, desirable properties inherent to cellulosic insulating materials should not be lost. For this, sufficient consideration should be given to urethane compounds which are to be employed for improving the heat stability of the cellulosic insulating materials. As a factor which might have detrimental effects on the desirable properties inherent to cellulosic insulating materials, there is considered the molecular weight of urethane compound employed. In case a urethane compound having a molecular weight of more than about 1000 is incorporated into a cellulosic insulating material, there is such a fear that the cellulosic insulating material would be injured in such inherent characteristics as permeability for insulating oils, resins or varnishes and would be deprived of softness (flexibility), which is peculiar to cellulosic insulating material. In case the molecular weight of urethane compound is less than about 1000, no such problems as mentioned above are brought about at all. What is of importance, therefore, is the upper limit of the molecular weights of urethane compounds employed. For this reason, there are selected in the present invention urethane compounds having a molecular weight of less than about 1000. Among the urethane compounds, even the lowest molecular weight compound has a molecular weight of 75. In the present invention, the use of urethane compounds low in molecular weight does not bring about any detrimental effect on cellulosic insulating materials at all, but serves to sufficiently achieve the objects of the present invention. For the above reason, the lowest urethane compound having a molecular weight of 75 is also useful for the present invention. According to the investigations, it was found that good results may be obtained in the case of compounds having a molecular weight of 100 to 300 because of economical and technical viewpoints.

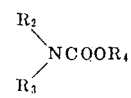
Urethane compounds usable in the present invention are as shown below.

1. Urethane compound defined by the general formula, NH_2COOR_1 (herein R_1 is either an aliphatic hydrocarbon group or aromatic hydrocarbon group).

These compounds include, for example:

NH_2COOCH_3
$NH_2COOCH_2CH_3$
$NH_2COOCH_2CH_2CH_3$
$NH_2COOCH(CH_3)_2$
$NH_2COOCH_2CH_2CH_2CH_3$
$NH_2COOCH_2CH{=}CH_2$
$NH_2COOC_6H_5$
$NH_2COOC_6H_4CH_3$
$NH_2COOC_6H_3(CH_3)_2$
$NH_2COOC_6H_4NO_2$

2. N-substituted urethane compounds defined by the general formula,



(wherein R_2 and R_3 are selected from the group consisting of aliphatic hydrocarbon groups, substituted aliphatic hydrocarbon groups, aromatic hydrocarbon groups, substituted aromatic hydrocarbon groups, chlorine atoms, hydrogen atom, and nitro group, and R_4 is an aliphatic hydrocarbon group, a substituted aliphatic hydrocarbon group, an aromatic hydrocarbon group, a substituted aromatic hydrocarbon group, a chlorine atom, or a nitro group.)

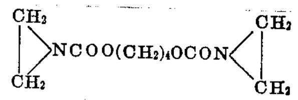
These compounds include, for example:

$HCONHCOOC_2H_5$
$C_3H_7CONHCOOCH_3$
$(CH_3)_2NCOOC_2H_5$
$C_2H_5NHCOOC_2H_5$
$C_6H_5NHCOOC_6H_4CH_3$
$NO_2NHCOOCH_3$
$CH_2ClNMCOOC_2H_5$
$ClNHCOOC_2H_5$
$Br_2NCOOC_2H_5$
$NO_2N(COOC_2H_5)_2$
$CH_3OCONHCOOCH_3$
$C_2H_5OCONHCOOC_2H_5$
$C_3H_7OCONHCOOC_3H_7$
$CH_3OCONHCOOC_2H_5$
$C_2H_5OCON(COOC_2H_5)_2$
$CH_3OCON(COOC_2H_5)_2$
$OONHCOO(C_2H_5)_2$
$CH_3CONHCOOCH_3$
$CH_3NHCOOC_2H_5$

3. Polyurethane compounds having at least two urethane bonds in the molecule thereof.

These compounds include, for example:

$C_2H_5OCONHCH_2NHCOOC_2H_5$
$C_6H_5OCONH(CH_2)_4NHCOOC_6H_5$
$CH_3OCONHC_{10}H_6NHCOOCH_3$
$C_2H_5OCONHC_{10}H_6NHCOOC_2H_5$
$C_2H_5OCONH(CH_3)C_6H_3C_6H_3(CH_3)NHCOOC_2H_5$
$H_2NCOO(CH_2)_2OCONH_2$
$CH_3NHCOO(CH_2)_2OCONHCH_3$



$CH_3(CH_2)_{17}OCONH(CH_2)_2$
$CH_3(CH_2)_{17}OCONH(CH_2)_2 \rightarrow N$
$CH_3(CH_2)_{17}OCONH(CH_2)_2$

75 – 1000 may be incorporated into a cellulosic insulating material.

The heat resistance-improving effect of the above-mentioned urethane compound (in the specification, heat resistance-improving effect also means heat stability of the cellulosic insulating material) is determined by the amount thereof incorporated into a cellulosic insulating material, and the amount of urethane compound to be incorporated varies depending on the amount of nitrogen contained in the compound. That is, the heat resistance-improving effect of urethane compound varies in substantial proportion to the amount of nitrogen incorporated into the cellulosic insulating material. However, if the amount of nitrogen incorporated is in excess of 5 percent by weight, said proportional phenomenon may be not be observed any more. In other words, when the amount of urethane compound calculated into the amount of nitrogen is up to 5 percent by weight, the heat resistance-improving effect of the urethane compound can be observed, but when said amount is more than 5 percent by weight, the heat resistance-improving effect reaches a saturation. On the other hand, the heat resistance-improving effect of urethane compound is scarcely observed if the amount of the compound calculated into the amount of nitrogen is less than 0.1 percent by weight. In the present invention, therefore, it is most desirable that the amount of the urethane compound, calculated into the amount of nitrogen, be selected from the range of 0.1 to 5.0 percent, and more particularly, of 0.3 to 2 percent, by weight so as to attain sufficient heat resistance with economical advantages.

When incorporated into a cellulosic insulating material, the urethane compound employed in the present invention and improve the heat resistance of said cellulosic insulating material. That is, the incorporation of the urethane compound increases the thermal stability of cellulosic insulating material.

For incorporation of the urethane compound employed in the present invention, there is adopted a suitable procedure. In case the urethane compound is in the form of liquid at normal temperature or is easily convertible to the form of liquid by heating, it can be directly incorporated into a cellulosic insulating material. Alternatively, a solution of the urethane compound, which has been formed by dissolving the compound in a suitable liquid medium, such as water, or an organic solvent, may be used to incorporate the urethane compound into a cellulosic insulating material. For example, the cellulosic insulating material is immersed in said solution of urethane compound and is then dried to remove the solvent, whereby the urethane compound can be incorporated into the insulating material. It is also possible to incorporate the urethane compound into a cellulosic insulating material by coating or spraying the insulating material with said solution of urethane compound to impregnate the insulating material with the solution, followed by drying. Alternatively, the object of incorporation of the urethane compound may be achieved in such a manner that one starting component of the urethane compound, which component is a hydroxyl group-containing organic compound selected from the group consisting of alcohols and glycols, is previously incorporated into a cellulosic insulating material and then the other starting component, which is an isocyanate compound, is incorporated into the insulating material, thereby reacting the two components to form the urethane compound in the cellulosic insulating material. Conversely, it is also effective to adopt such procedure that the isocyanate compound is first incorporated into the insulating material and then the alcohol or glycol is incorporated, thereby reacting the two components to form the urethane compound in the cellulosic insulating material. In some cases, the urethane compound may be incorporated into the cellulosic insulating material at the time of application of the insulating material to electrical apparatuses or at the time of making the cellulosic insulating materials.

The aforesaid suitable solvent means a solvent which is chemically stable and which does not denature cellulosic insulating materials. Preferable as such solvents include aliphatic and aromatic organic solvents such as alcohols, acetone, methylethylketone, ether, benzene, toluene, xylene and the mixtures thereof. In addition thereto, water and the like are considerably useful, as well. Of course, water and the above mentioned organic solvent may be used as a mixed solvent.

The urethane compounds employed in the present invention have particularly excellent heat resistance-improving effects on such cellulosic insulating materials as papers, e.g. kraft paper and Manila paper; craped kraft and Manila papers; cotton cloths; cotton cloth tapes; wood; pressboards; or fiber sheets. These cellulosic materials are made of natural raw material, of course, from viewpoints of low costs thereof.

The cellulosic insulating materials in accordance with the present invention are applicable to various electrical equipment.

FIG. 1 is a drawing showing one application example of the present cellulosic insulating material and is a partial longitudinal section of a power transformer. In FIG. 1, **1** is an oil tank, **2** is an iron core, **3** is a low tension winding, **4** is a high tension winding, **5** is a barrier insulation between the coils (main insulation), and **6** is an insulating oil. In the transformer shown in FIG. 1, the cellulosic insulating material of the present invention has been applied to layer insulations of the windings **4**, **5**. In the case of a transformer of the like, the cellulosic insulating material employed for the layer insulations are, in general, kraft paper, Manila paper, craped kraft paper, craped Manila paper or cotton cloth tape. In such a case, therefore, there is used a cellulosic insulating material prepared by incorporating the aforesaid urethane compound into the above-mentioned insulating paper or cotton cloth tape. Further, the present cellulosic insulating materials are useful as insulating materials between barrier insulation **5** of the high tension and low tension windings.

Further, the cellulosic insulating materials of the present invention are effectively applied to the case as shown in FIG. 2. FIG. 2 is a rough cross-sectional sketch of the stator slot portion of a rotating machine. In FIG. 2, **7** is a stator, **8** is a stator slot, **9** shows stator coils, 10 is a stator slot liner, 11 is an insulating material disposed between the layer insulations of stator coils 9, and 12 is a slot wedge. Into the stator slot 8 are inserted the stator coils 9, which are insulated from the stator 7 by means of the stator slot-liner 10. Further, between the layer insulations of the stator coils 9, there is also employed the insulating material 11. As said stator slot liner 10 and said insulating material 11 between the layer insulations of stator coils 9, the cellulosic insulating materials of the present invention can be used, as well. The stator coils 9 of this kind of rotating machine are impregnated with a resin, in most cases. Even in such cases, the cellulosic insulating materials of the present invention are effective. Thus, the cellulosic insulating materials of the present invention are effective as insulating materials not only for oil filled electrical apparatuses and rotating machines but also for so-called dry type electrical apparatuses such as dry type transformers.

Properties of the present cellulosic insulating materials, which are utilized in the manner as mentioned above, are apparent by examining the polymerization ratio of said cellulosic insulating materials and the characteristics or behaviors after heat degradation of other co-existing insulating materials, and the tan δ values of the cellulosic insulating material. These are shown in FIGS. 3 to 7.

Now, FIGS. 3 to 7 will be illustrated below along with embodiments of the present invention.

Samples employed in the examination of characteristics of the present cellulosic insulating materials were prepared in the following manner:

The urethane compounds set forth in Table 1 were individually dissolved in ethanol. Insulating papers were immersed in the resulting solutions to impregnate said papers with the solutions of said urethane compounds, and were then dried at room temperature. After drying, the papers were conditioned for more than 3 days in an atmosphere at a relative humidity of 66 percent.

The thus prepared samples were subjected to a heat degradation test (accelerated thermal aging test, in another words) under somewhat severe conditions as mentioned below as a means to effect accelerated degradation.

That is, each sample (urethane compound-incorporated cellulosic insulating paper) was charged into a stainless steel autoclave and was heated for 24 hours in a nitrogen gas atmosphere under such conditions as a temperature of 175°C and a pressure of 15 atm. The heat resistance of each sample was evaluated according to a process best suitable for evaluation of heat degradation, i.e. a process carried out by measuring the polymerization ratio of the sample cellulosic insulating paper and calculating the percentage of the polymerization ratio thereof after heat degradation to the initial polymerization degree, namely the polymerization ratio. The results are as set forth in Table 1, in which the test result of a non-treated kraft paper (a standard kraft paper) is also shown for comparison in order to indicate the extent of heat resistance-improving effects of the urethane compounds.

TABLE 1

Urethane compound	Incorporated amount (wt. %)	Polymerization ratio (%)
Non-treated kraft paper	—	25
$NH_2COOCH_2CH_2CH_2CH_3$	0.54	78
$CH_3CONHCOOCH_3$	0.65	68
$CH_3NHCOOC_2H_5$	0.70	76
$C_6H_5NHCOOC_2H_5$	1.02	70
$C_2H_5OCONHCOOC_2H_5$	0.71	71
$C_2H_5OCONHCH_2NHCOOC_2H_5$	0.84	66
$CH_3OCONHC_{10}H_6NHCOOCH_3$	0.59	62
$NH_2COO(CH_2)_2OCONH_2$	1.32	78

(In Table 1, the incorporated amount is the amount of nitrogen in the urethane compound incorporated into the cellulosic insulating material.)

From Table 1, it is evident that the heat resistance-improving effects of the urethane compounds are marked.

FIG. 3 shows the results of investigation in heat resistance of samples in an insulating oil, and is a graph showing variations in polymerization ratio of said samples when heat degraded by immersing them in an insulating oil at 175°C. The sample insulating papers employed in the above case were a urethane compound-incorporated insulating paper A in accordance with the present invention, a conventional urea-incorporated insulating paper B, a tetraethylene-pentamine-incorporated insulating paper C, and a non-treated insulating paper (standard kraft paper) D. In view of the curve of the present urethane compound-incorporated insulating paper A, it is clear that said paper A is more excellent in heat stability than the urea-incorporated insulating paper B, which is particularly excellent in heat stability among the conventional insulating papers.

In the next place, a cellulosic insulating paper incorporated with the urethane compound $NH_2COO(CH_2)_2OCONH_2$ bis-(methylcarbamate) and a non-treated cellulosic insulating paper were subjected to a heat degradation test by heating them in a closed vessel together with a magnet wire, an insulating oil and a core. That is, given amounts of each sample insulating paper, the magnet wire and the core were placed in a glass test vessel and were heated under reduced pressure at 105°C for 16 hours. Thereafter, an insulating oil, which had been degasified and dehydrated under reduced pressure, was charged into said vessel, and air was sealed in the upper portion of the vessel. Subsequently, the vessel was heated at 175°C for 20 days to degrade the above-mentioned materials. After the heat degradation, the polymerization ratio of the sample insulating papers, the breakdown voltage of the insulating film of the magnet wire, the dielectric dissipation factor (tan δ) and volume resistivity (Ω-cm) of the insulating oil were measured to obtain the results as shown in Table 2.

TABLE 2

	Item											
	Polymerization ratio (percent)			Breakdown voltage of insulating film of magnet wire (kv.)			Dielectric dissipation factor of insulating oil tan δ (percent).			Volume resistivity of insulating oil ($\times 10^{13}$ Ω—cm.)		
Days of heating	Initial	10 days	20 days	Initial	10 days	20 days	Initial	10 days	20 days	Initial	10 days	20 days
Incorporated amount (wt. percent):												
0.5	800	310	230	11.2	9.4	6.6	0.02	0.06	0.09	150	6.1	5.9
1.32	800	350	280	11.2	10.6	8.6	0.02	0.06	0.10	150	12.0	7.5
Non-treatment	800	120	<100	11.2	9.7	2.6	0.02	0.08	0.12	150	11.0	1.9

NOTE.—In Table 2, the incorporated amount is the amount of nitrogen in the urethane compound incorporated into the cellulosic insulating paper.

In view of Table 2, it is understood that the cellulosic insulating paper according to the present invention is extremely low in degradation of various properties due to thermal influence and has little detrimental effect on other insulating materials present together therewith. When the case where the incorporated amount of urethane compound is 0.5 percent by weight is compared with the case where said amount is 1.32 percent by weight, no deterioration in properties of the insulating film of magnet wire and of the insulating oil is observed. And, the case where the incorporated amount of urethane compound is greater gives favorable results to the properties of co-existing other insulating materials such as the insulating film of magnet wire or the insulating oil. This is readily understandable also from FIG. **4**. FIG. **4** shows the results of a similar heat degradation test to that mentioned above which was carried out in order to calculate the adhesive strength ratio of a magnet wire (polyvinylformal enamel wire). (The number of twists of the enamel wire is the number of twists attained in the case where one end of the enamel wire is fixed and the other end is twisted until the enamel film peels.) The adhesive strength ratio referred to herein is the percentage of the number of twists of the enamel wire after heat degradation to that of the wire before heat degradation (i.e. number of twists after heat degradation × 100/initial number of twists). According to FIG. **4**, it is clear that among the cases where conventional cellulosic insulating papers F, G and H are used, only the case of urea-incorporated insulating paper G has favorable efficiencies equivalent to the case of the cellulosic insulating paper E of the present invention, and that in the cases of non-treated insulating paper F and tetraethylene-pentamine-incorporated insulating paper H, there is observed marked degradation in adhesive strength of enamel insulating film, which is a co-existing insulating material. From this, it is understood that in case the cellulosic insulating paper E of the present invention is used, there is no such fear that other co-existing insulating materials are adversely affected. That is, the urethane compounds, which are incorporated according to the present invention into cellulosic insulating materials, have such advantages that they impart favorable results to other co-existing insulating materials but do not cause undesirable results therewith.

From FIG. **5** also, it is understood that when the cellulosic insulating materials of the present invention are used, the degradation of other cellulosic insulating papers and insulating oils, which are used together with the present insulating materials, is far less than in the case of conventional insulating materials. FIG. **5** shows the results of heat degradation tests carried out under the same conditions as above to measure the variations in acid values of insulating oil employed in the tests together with insulating papers I, J, K and L, and is a graph showing the relationship between the days of heat degradation and the acid value of said insulating oil. From the graph shown in FIG. **5**, it is clear that in case the cellulosic material I of the present invention is used, the degree of increase in acid value of the insulating oil is lower than in the case of conventional non-treated insulating paper J, urea-incorporated insulating paper K and tetraethylenepentamine-incorporated insulating paper L. This indicates that when the present cellulosic insulating material is used, the amounts of acid substances formed by heat degradation of the co-

existing insulating materials, particularly cellulosic insulating paper and insulating oil, are small. In other words, the urethane compound may catch and neutralize the acid substances so that the heat degradation of the cellulosic insulating material is restrained sufficiently.

In FIG. **6**, the dielectric dissipation factor of cellulosic insulating paper M in accordance with the present invention are shown in comparison with those of conventional insulating papers N, P and Q. The measurement of dielectric dissipation factor was effected after placing each sample insulating paper on an aluminum foil, rolling the resulting composite to prepare a condenser element and impregnating the condenser element with an insulating oil. From FIG. **6**, it is understood that the present cellulosic insulating paper M is equivalent in dielectric dissipation factor values to the non-treated insulating paper N, which has been known to be low in said value, and is far lower in said value than the conventional cellulosic insulating papers, i.e. urea-incorporated insulating paper P and tetraethylene-pentamine-incorporated insulating paper Q.

Further, the present and conventional cellulosic insulating materials were subjected to moterette test (AIEE, No. **510**), which has most widely been adopted as a method for measuring heat resistance of cellulosic insulating materials, to obtain the results as shown in FIG. **7**. From these results also, it is understood that the present cellulosic insulating paper R is more excellent in heat resistant life than the conventional non-treated insulating paper S and urea-incorporated insulating paper T. Since there is such a fear that in the case of common oil-filled electrical apparatuses, the urethane compounds incorporated into cellulosic insulating materials might elute and migrate into the insulating oils to lower the effects of improving the heat resistance of the cellulosic insulating materials, it is desirable that the oil solubility of the urethane compounds be small. In view of the above, cellulosic insulating papers incorporated, respectively, with $NH_2COOCH_2H_2CH_3$ and $NH_2COOCH_2CH_2CH_2CH_3$ were individually immersed in an insulating oil and were heated for about 2 months at 130°C, which is somewhat higher than the temperature at which general electrical apparatus is used, and then the amounts of the urethane compounds remaining in the insulating papers were measured. The results are shown in Table 3. The amounts of remaining urethane compounds are represented by values calculated into the amounts (wt. %) of nitrogen in the urethane compounds.

TABLE 3

Urethane compound incorporated into cellulosic insulating paper	Amount of nitrogen in cellulosic insulating material (wt.%)	
	Before heating	After heating
$NH_2COOCH_2CH_2CH_2CH_3$ (n-butyl carbamate)	0.69	0.64
$NH_2COOCH_2CH_2CH_3$ (iso-propyl carbamate)	0.48	0.47

The above results show that even the urethane compounds relatively low in molecular weight have scarcely eluted and migrated into the insulating oil, the therefore it may be considered that there is no fear of elution and migration either in the case of other large portions of urethane compounds.

What is claimed is:

1. An electrical apparatus operated at a temperature above room temperature which comprises:

a. electrical conducting means;

b. a cellulosic insulating material wrapped on said electrical conducting means; and

c. an insulating oil being in contact with said cellulosic insulating material,

said cellulosic insulating material containing at least one urethane compound having a molecular weight of from 75 to 1000 in an amount of from 0.1 to 5 percent by weight of said cellulosic insulating material, based on the amount of nitrogen contained in said urethane compound, so that deterioration of said cellulosic insulating material is suppressed.

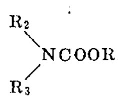
2. An electrical apparatus according to claim 1, wherein said amount of said urethane compound is from 0.3 to 2 percent by weight, based on the amount of nitrogen.

3. An electrical apparatus according to claim 1, wherein said urethane compound has a molecular weight of from 100 to 300.

4. An electrical apparatus according to claim 1, wherein said urethane compound is selected from the group consisting of

1. urethane compounds having the formula NH_2COOR_1 wherein R_1 is an aliphatic hydrocarbon group or an aromatic hydrocarbon group;

2. N-substituted urethane compounds having the formula



wherein R_2 and R_3 are each an aliphatic hydrocarbon group, a substituted aromatic hydrocarbon group, chlorine, hydrogen or nitro and R_4 is an aliphatic hydrocarbon group, a substituted aliphatic hydrocarbon group, an aromatic hydrocarbon group, a substituted aromatic hydrocarbon group, chlorine or nitro; and

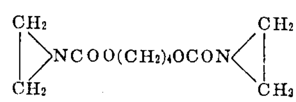
3. polyurethane compounds having at least two urethane bonds in the molecule thereof.

5. An electrical apparatus according to claim 1, wherein said cellulosic insulating material comprises at least one member selected from the group consisting of kraft paper, Manila paper, craped paper, cotton cloth, wood and press-board.

6. An electrical apparatus according to claim 4, wherein said urethane compound comprises at least one member selected from the group consisting of NH_2COOCH_3, $NH_2COOCH_2CH_3$, $NH_2COOCH_2CH_2CH_3$, $NH_2COCH(CH_3)_2$, $NH_2COOCH_2CH_2CH_2CH_3$, $NH_2COOCH_2H{=}CH_2$, $NH_2COOC_6H_5$, $NH_2COOC_6H_4CH_3$, $NH_2COOC_6H_3(CH_3)_2$, and $NH_2COOC_6H_4NO_2$.

7. An electrical apparatus according to claim 4, wherein said urethane compound comprises at least one member selected from the group consisting of $HCONHCOOC_2H_5$, $C_3H_7CONHCOOCH_3$, $(CH_3)_2NCOOC_2H_5$, $C_2H_5NHCOOC_2H_5$, $C_6H_5NHCOOC_6H_4CH_3$, $NO_2HCOO(C_2H_5)_2$, $CH_2ClNHCOOC_2H_5$, $Br_2NHCOOC_2H_5$, $CH_3OCONHCOOCH_3$, $C_2H_5OCONHCOOC_2H_5$, $C_3H_7OCONHCOOC_3H_7$, $CH_3OCONHCOOC_2H_5$, $CH_3CONHCOOCH_3$ and $CH_3NHCOOC_2H_5$

8. An electrical apparatus according to claim 4, wherein said urethane compound comprises at least one member selected from the group consisting of $C_6H_5OCONH(CH_2)_4NHCOOC_6H_5$, $CH_3OCONHC_{10}H_6NHCOOCH_3$, $H_2NCOO(CH_2)_2OCONH_2$, $CH_3NHCOO(CH_2)_2OCO(NHCH_3)_3$, $C_2H_5OCONHCH_2NHCOOC_2H_5$ and



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