May 26, 1970 M. G. ROCHOLL ET AL 3,514,581

OPTICALLY TRANSPARENT ELECTRICAL HEATING ELEMENT

Filed May 29, 1968

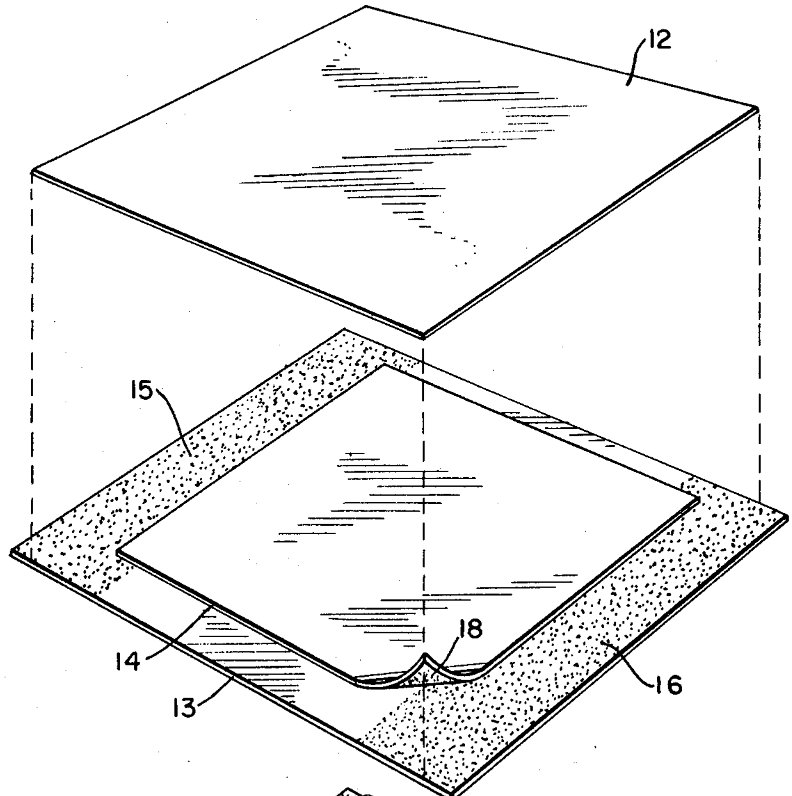


FIG. 1

FIG. 2

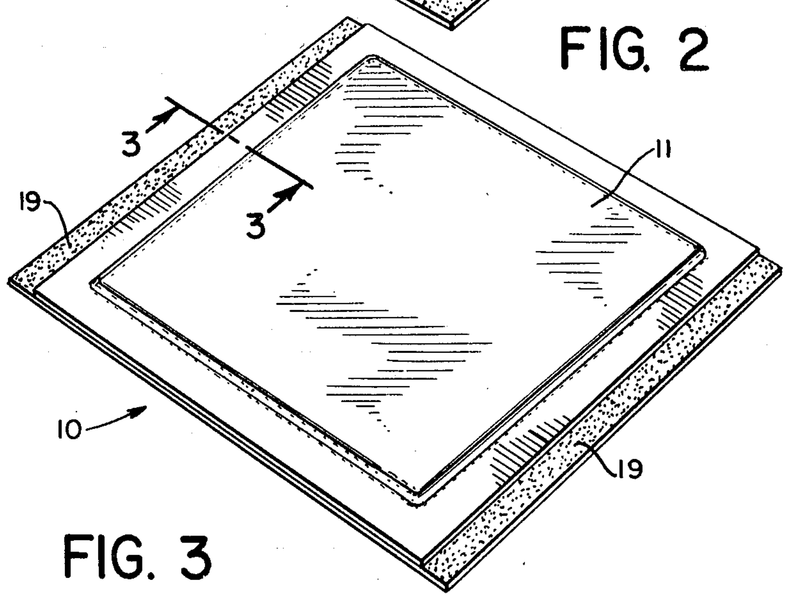
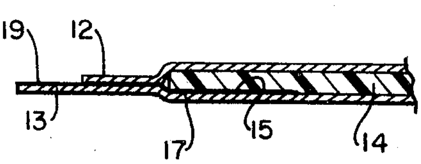


FIG. 3



INVENTORS
MARTIN G. ROCHOLL
ILSE ROCHOLL
BY Darby & Darby
ATTORNEYS

United States Patent Office

3,514,581

Patented May 26, 1970

3,514,581
OPTICALLY TRANSPARENT ELECTRICAL HEATING ELEMENT

Martin G. Rocholl and Ilse Rocholl, Heidelberg, Germany, assignors to Gulton Industries, Inc., Metuchen, N.J., a corporation of New Jersey

Filed May 29, 1968, Ser. No. 733,050
Claims priority, application Austria, June 2, 1967, A 5,128/67
Int. Cl. H05b *3/50, 3/34*
U.S. Cl. 219—522 18 Claims

ABSTRACT OF THE DISCLOSURE

An optically transparent heating element is described which comprises an electrolyte solution preferably absorbed on an optically transparent support material having electrical conductors in electrical contact relationship with electrical conductors of an optically transparent insulation sheet which is stable to the electrolyte solution, the electrolyte support material being sealed within the optically transparent insulating sheet. The optically transparent heating element has a positive temperature coefficient and is self-regulating in that it does not have to be controlled by interrupting the current, but adjusts itself automatically to limit its temperature in individual areas thereof according to the heat load in such individual areas by automatically varying its local resistivity in such areas.

This invention relates to an optically transparent, self-regulating heating element.

Heating elements known in the prior art are generally composed of metal wires, foil, carbon, carbon-containing materials, semiconductors (e.g. silicon carbide) and the like. These heating elements find diversified use in such consumer goods as cooking appliances, heating apparatus, lighters, etc. In the case of metal and semiconductor heating elements, current transfer is effected by electron conduction. Such heating elements absorb the electromagnetic energy of light, and as a result are either opaque or intensely colored. Similarly, organic semiconductors which work on the principle of electron conduction are also intensely colored substances.

Hence the foregoing type of heating elements are not suitable for use where optical transparency is desired or necessary such as in windows of automobiles, homes, offices, etc. Such windows are a considerable source of heat absorption during cold weather which frequently results in discomfort to the occupant of an otherwise properly heated enclosure. Moreover, a serious problem occurs where condensation forms on any window surface of a vehicle where maximum optical transparency is essential. A reduction in visibility whether due to ice, snow or condensation on a window surface is a serious driving hazard. Therefore, there is a great need for a heating element which prevents condensation on a window surface by maintaining the window surface at an appropriate temperature and which will maintain the inside surface of a window at the same temperature as other non-transparent portions of an enclosure so as to substantially eliminate heat absorption by the window surface. Such a heating element must be optically transparent and is provided by the present invention.

Accordingly, it is an object of the present invention to provide an optically transparent electrical heating element in the form of a flexible or rigid sheet or other shaped article that uses the ions produced by an electrolyte solution as the electric current carrier.

A further object of the present invention is to provide an optically transparent heating element which comprises an inorganic and/or organic acid, base and/or salt, dis-

solved in an aqueous or non-aqueous solvent to furnish an electrolyte solution which is enclosed by an insulating sheet material provided with conductive means for passing an electric current through the electrolyte solution.

Yet another object of the present invention is to provide an optically transparent electrical heating element which comprises an electrolyte solution absorbed on an optically transparent support material and an optically transparent insulating material which encases the electrolyte-containing support material and provides an airtight seal around the support material, the support material and insulating material having electrical conductors in electrical contact relationship to provide for current flow inside the sealed portion of the optically transparent heating sheet containing the electrolyte support material.

An additional object of the present invention is to provide an optically transparent heating element having a positive temperature coefficient which does not require the heating element to be controlled by interrupting the current supply, as by means of a thermostat or the like, but which adjusts itself automatically to limit its temperature in individual regions thereof according to the heat load in such individual regions by automatically varying the electrical resistivity in such regions.

Other objects and advantages of the present invention will become apparent from the following description and the annexed drawings, in which:

FIG. 1 is an exploded perspective view of the components of an optically transparent heating element manufactured in accordance with the present invention;

FIG. 2 is a perspective view of an assembled optically transparent heating sheet according to the present invention; and

FIG. 3 is a fragmentary view along lines **3**—**3** of FIG. 2.

In accordance with the present invention an optically transparent electrical heating element, preferably in the form of a relatively flat sheet, is provided which consists of an aqueous or non-aqueous electrolyte solution contained within an optically transparent insulating envelope which is stable and inert toward the electrolyte and which is provided with conductive means for passage of electrical current, as will be explained with reference to the drawing.

The term "sheet" as used herein includes flexible or rigid foil, film, plate, etc. The term "insulating envelope" includes two relatively flat, flexible or rigid sheets joined together so as to provide a sealed compartment.

When a flexible electrical heating element is desired, the electrolyte solution is preferably used in conjunction with an optically transparent support material in the form of a relatively flat sheet which has the characteristic of being able to support the electrolyte without itself going into solution as by absorption, adsorption, or in any other way. This support material with the electrolyte will hereinafter be referred to as the "electrolyte sheet." The support material is preferably a polymeric substance such as a plastic, rubber or protein material. The polymeric substance is preferably cross-linked in accordance with procedures well known in the art.

An optically transparent electrical heating element according to the present invention is illustrated in the drawings. Referring to FIG. 2, there is shown a heating element **10** comprising an envelope portion **11** having a pair of opposing side wall portions **12** and **13** shown in FIGS. 1 and 3, each overlying one surface of a relatively flat electrolyte sheet **14** which supports the electrolyte. The side wall portion **12** is of smaller dimension than side wall portion **13**, as shown in FIGS. 1 and 3 to leave protruding conductive strips **19** on side wall portion **13**. Side wall portion **13** is provided with electrodes **15** and **16** along two opposed edges and the electrolyte sheet is also provided with electrodes **17** and **18** along two opposed edges so that electrodes **15** and **17** will be in overlying and electrical contact relationship when the heating element is assembled. Similarly, electrodes **16** and **18** will be in the same overlying and electrical contact relationship to each other in the assembled heating element as electrodes **15** and **17**. Electrodes **15**, **16**, **17** and **18** may comprise conductive silver in the form of a thin foil having an adhesive coated on one surface for securing the foil to the support material. The selected adhesive must be insoluble in the electrolyte that is to be absorbed on the support material. Rather than foil the electrode may be a powder containing an adhesive which is coated onto the support material or conductive wires secured to the support material at its opposed edges or incorporated therein along the edges in a suitable manner. Other materials having a high polarization voltage such as platinum, nickel and amalgamized silver may be used to form the electrodes.

The heating element **10** is assembled by placing electrolyte sheet **14** between side wall portions **12** and **13** and then sealing the edges of these side wall portions in a conventional manner such as with a high frequency welder or other hot sealing device. When side wall portions **12** and **13** are sealed, the envelope portion **11** is formed which encases electrolyte sheet **14**. Conductive section of each of conductive electrodes **15** and **16** which comprise a foil of silver adhesively secured to side wall portion **13** extends outside of the envelope portion **11** and the remaining portion of each of electrodes **15** and **16** extends within the envelope portion **11** and in overlying electrical contact relationship with conductive silver electrodes **17** and **18** which comprise a foil of silver adhesively secured on electrolyte sheet **14**. This arrangement permits passage of current from outside of envelope portion **11** of heating element **10** into the envelope portion through leads (not shown) which are connected to the exposed section of electrodes **15** and **16** and to an alternating current supply source (not shown).

Changes may be made in the construction without departing from the spirit of the invention. It is to be understood that the foregoing description and drawing have been given primarily to facilitate an understanding of the invention and should be interpreted as illustrative only.

Electrolytes which are suitable for use with the present invention comprise aqueous or non-aqueous solutions of (a) inorganic acids, (b) organic acids, (c) inorganic and organic bases and/or salts, as well as combination of the foregoing which dissociate into ions in the aqueous or non-aqueous solvent. An aqueous electrolyte may comprise, for example, materials such as hydrochloric acid, potassium chloride, phosphoric acid, acetic acid, boric acid, formic acid, etc., or a mixture of an acid and salt such as phosphoric acid and potassium phosphate, acetic acid and sodium acetate or buffer mixtures dissolved in water. The selection of a particular inorganic or organic acid is not critical except that highly oxidizing acids are not desirable nor is hydrogen fluoride suitable.

Non-aqueous electrolytes are obtained by dissolving a dehydrated inorganic and/or organic acid, base and/or salt in a non-aqueous solvent. The selected electrolyte must be one which will form ions in the desired non-aqueous solvent. Examples of suitable non-aqueous solvents include propylene carbonate, dimethyl formamide, dimethyl sulfamide, acetonitrile, nitrobenzene, formamide, lactic acid, N-ethyl-morpholine, monoethanolamine, ethyl acetate, cyclohexanone, 2,4-pentadione, 2-pentanone, tetramethylene sulfones, 2-chloroethanol, ethylene dipropionate, epichlorohydrin, furfural, methylthiocyanate, propionitrile, acrylonitrile. Other non-aqueous solvents may also be employed. Examples of salts that may be used with non-aqueous solvents include lithium perchlorate, sodium iodide, potassium iodide, etc.

The selection of a suitable support material is not critical except to the extent that it is desirably optically

transparent and should be compatible with the electrolyte solution. This means that the selected support material must be capable of supporting the selected aqueous or non-aqueous electrolyte solution without dissolving in that solution. As an example, when employing an aqueous electroltye in conjunction with a support material, the support material may be one which absorbs water or swells in water. Examples of such support materials include hydrocellulose, methyl cellulose, cellulose, carboxyl methyl cellulose, gelatin, agar agar, pectins, starch, dextrins, gum arabic, aginates, hemicellulose and the like.

Where the electrolyte is a concentrated acid or base which is dissociated in water, suitable support materials include cellulose, acetate, cellulose acetobuylate, cellulose propionate, ethyl cellulose, artificial horn, phenol molding resin, polyacetates, polyamides, polycarbonate, celluloid and polyesterpolyurethane elastomers. When the electrolyte is dissolved in a non-aqueous solvent such as an aliphatic and/or aromatic hydrocarbon of the type heretofore exemplified, the optically transparent support material may be selected from such materials as natural rubber, polyisobutylene, polymethacrylate, polystyrene, acrylonitrile-styrene copolymers, butadiene-styrene copolymers, polyvinyl carbozol, polyvinylchloride (hard and saft containing about 40% plasticizer), ethyl cellulose, cellulose acetobutyrate, cellulose propionate, ABC resins (e.g. blend of a butadieneacrylonitrile copolymer with polystyrene). Where the non-aqueous solvent for the electrolyte is an alcohol, ester, ketone, ether or haloalkane, the following are exemplary of optically transparent polymeric support materials: ethyl cellulose, cellulose acetate, cellulose acetobutyrate, cellulose propionate, celluloid, polyester resins, polycarbonate, polyisobutylene, polymethyl methacrylate, polyvinylchloride (hard and soft) and poyester-polyurethane elastomers.

The optically transparent electrolyte sheet used in the heating element of the present invention may be produced as follows: a polymeric support material in the form of a relatively flat sheet is selected which is compatible with the electrolyte solution that is to be used and provided with opposed spaced conductive electrodes **15** and **16** as previously described prior to introduction of the electrolyte solution into the sheet.

To determine the necessary spacing between the electrodes for a desired heat performance (watts/square meter) of the heating element, a small strip of the selected support material is placed in the desired electrolyte solution to effect swelling. After removal from the electrolyte solution the support materials square resistance is measured. This square resistance depends on the layer thickness of the support material, its swelling capacity in the electrolyte solution, the electric conductivity of the electrolyte and the swelling time of the support material in the electrolyte. After the square resistance has been measured, the electrode spacing for the desired heat output and the available working voltage acn be readily calculated.

After the electrodes are secured to the selected support material, this material is placed in the electrolyte solution for a time period which may range anywhere from 10 seconds to about 4 hours, depending on the absorbing and swelling characteristics of the material as well as the square resistance which is desired and on which the electrode spacing is based. The swelling of the support material may be carried out at room temperature or at elevated temperatures. The selection of a temperature is not critical and the temperature may be adjusted in accordance with the absorption properties exhibited by the support material at different temperatures.

After the desired degree of absorption has been reached, the excess electrolyte on the surface of the electrolyte sheet is removed by wiping the sheet with a rubber wiper or the like, leaving the electrolyte sheet damp but not wet. It is important not to dry out the electrolyte sheet because this would adversely affect current flow in the heating element.

As an alterntive to placing the support material in the electrolyte solution to produce the electrolyte sheet, it is possible, where a non-aqueous electrolyte solution is being used, to achieve absorption or swelling by first subjecting the support material to heated vapor of the pure solvent of the electrolyte and thereafter placing the support material in the electrolyte solution.

The insulating envelope in which the electrolyte sheet is encased should meet the following criteria: (1) desirably optically transparent, (2) stable toward the electrolyte in the electrolyte sheet, i.e. does not absorb the electrolyte, and (3) substantially impervious to air and to the electrolyte solvent. The selection of the insulating material is governed to a large extent by the nature of the electrolyte selected for the heating element. For example, the following flexible insulating materials are suitable for use with non-oxidizing aqueous electrolyte solutions: chlorinated polyether, polyethylene, polystyrene, polyvinylcarbazol, "Teflon," polytrifluorochloroethylene, etc. Where the electrolyte is dissolved in an aliphatic and/or aromatic solvent, alcohol, ketone, ether, ester or haloalkane, insulating materials that are suitable include epoxy resins, urea-resin molding compounds, artificial horn, melamine resin molding compounds, phenol resin molding compounds, polyacetate, chlorinated polyether, polyamides, "Teflon," polytrifluorochloroethylene, "Vulcanfiber" and polyester-polyurethane elastomers. Where the electrolyte is a strong acid suitable insulating materials include chlorinated polyethers, polyethylene of high or low density, polyisobutylene, polymethyl methacrylate, polypropylene, polystyrene, polyvinylchloride (hard and soft) "Teflon" and polytrifluoroethylene. Insulating materials other than those exemplified above may also be employed, depending on the nature of the electrolyte material. For example, some polymeric materials are not stable in acid solutions but are stable toward basic solutions. Therefore, if the electrolyte comprises a base solution material such as a styrene-acrylonitrile copolymer, a styrene-butadiene copolymer, ABS resins, polyvinyl carbazol, etc. may be employed because they are stable toward bases but not acids. The thickness of the insulating sheet is preferably in the range from about 0.1 mm. to about 1.0 mm.

While the foregoing are primarily flexible sheet materials, rigid transparent insulating sheets, e.g., glass plates, are also suitable. In such case, it is not necessary to use an electrolyte support sheet, and the electrolyte may be in liquid form between two flat close spaced rigid sheets.

The following examples are illustrative of how to produce an optically transparent electrical heating element in accordance with the present invention.

EXAMPLE 1

An electrolyte solution was prepared by dissolving 2 moles of potassium chloride in one mole hydrochloric acid.

A test strip of hydrocellulose (cellulose of the formula $(C_6H_{10}O_5)_{200-450}$ treated with NaOH to obtain a smooth and gleaming surface) having a length of 10 cm., a width of 4 cm., and a thickness of 0.03 mm. was provided with conductive silver foil along two of its edges, the spacing being 8 cm. This test strip was placed in the electrolyte solution at 20° C. for two minutes which resulted in swelling of the test strip due to absorption of the electrolyte. The test strip was removed from the electrolyte solution, and its dimensions again measured. The thickness had increased to 0.062 mm. and the length and width increased about 10%. The square resistance of this test strip was 1030 ohms.

Based on the information obtained from the swelled test strip, a sheet of dry hydrocellulose foil having a thickness of .03 mm. was provided with conductive silver electrodes in the form of a thin foil having a width of

5 mm. along two edges and placed in the electrolyte solution at 20° C. for two minutes. The geometric dimensions of the resulting swelled foil are 26 cm. x 15 cm. x .062 mm. The conductive silver electrode spacing along the edges of the swelled foil was 25 cm.

This swollen optically transparent support material is placed between two sheets of optically transparent polyethylene having a thickness of about .01 mm. which has been provided with conductive silver foil electrodes along two of its edges as illustrated in the drawing. The electrodes are spaced 25 cm. apart and the width of each electrode is 2 cm. The two sheets are hermetically sealed by heat along the edges as illustrated in the drawing.

This optically transparent heating element was found to have a square resistance of 1100 ohms and an operating temperature of about 48° C. at room temperature.

EXAMPLE 2

An optically transparent heating element was manufactured according to Example 1, with the presence of 10% by weight of glycerine in the electrolyte solution. The quantity of glycerine is not critical but is preferably in the range of 5 to about 20% by weight.

EXAMPLE 3

An optically transparent heating element was prepared in accordance with Example 1, using hydrocellulose foil and an electrolyte which is an aqueous solution of boric acid.

While it is not desired to be limited to any theories of operation, it is believed that the following explanation of how the optically transparent heating element of the present invention provides a self-regulating heating element will aid in undertanding and appreciating the present invention. As previoussly indicated, the optically transparent heating element preferably has a positive temperature coefficient.

To obtain controlled heating from the optically transparent heating element of the present invention, the current absorption of any individual area of he heating element is preferably made self-regulating. This means that those areas or zones of the heating element will use less current where there is an increase in resistance due to an increase in temperature in the zone and hence self-regulate its temperature, whereas relatively colder areas of the heating element heat up faster due to a decreased electrical resistance and greater current consumption.

It is known that upon heating an electrolyte solution, electrolyte ion mobility and hence electrical conductivity increases sharply initially as a result of the decrease in viscosity of the electrolyte solution. However, as the temperature of the heated electrolyte solution continues to rise, the speed of migration of the ions decreases due to the strong thermal movement of the solution molecules which counteracts the initial increase in ion migration. As a result of this phenomena a positive temperature coefficient can be achieved by either (a) creating a temperature-dependent equilibrium between (i) the vapor pressure increase within the sealed optically transparent heating element due to increased temperature in an individual region of the heating element and (ii) solvent reabsorption by the electrolyte sheet upon cooling in the same region of the heating element or (b) through the use of a non-aqueous solvent for the electrolyte that has a pronounced temperature coefficient of the dielectric constant which over-compensates for the sharp conductivity increase initially upon heating of the electrolyte resulting from the decrease in viscosity of the solvent.

Generally, the temperature-dependent equilibrium is achievable with electrolytes whose solvent possesses within the working temperature range (i.e. between about 35° C. and about 80° C.) of the heating element of the present invention a sufficiently high vapor pressure, so that the solvent can evaporate from the electrolyte sheet in the envelope portion of the heating element. Generally, solvents with boiling points of less than about 120° C.

(e.g. aqueous solutions of salts, acids or bases diluted with water) have a sufficiently high vapor pressure so that the solvent will evaporate from the electrolyte sheet. To assure that the vaporization does not result in complete drying out of the electrolyte sheet, when using an aqueous electrolyte and thereby produce current cut-off, a highly hydroscopic compound, e.g. magnesium perchlorate, concentrated phosphoric acid, etc., may be included in the electrolyte solution. Such a compound will also accelerate reabsorption of the evaporated solvent in those regions of the heating element which are cooling.

Therefore, when the electrolyte solvent has a high vapor pressure within the working temperature range of the heating element, a reduction of the heat dissipation at a particular region of the heating element, such as by placing an object in front of such region of the heating element, will initially result in an increase in temperature in that area. This increase in temperature will result in a rise in temperature of the electrolyte solvent thereby promoting its vaporization. As a result of the increased vaporization of the electrolyte in the blocked region, there is a corresponding increase in electrical resistance which is due to a decrease in ion mobility in those areas where there has been a vapor pressure increase. This decrease in ion mobility produces an increase in resistivity which results in decreased current flow in the blocked region.

Thus, an optically transparent heating element, one square meter which is blocked in one region would exhibit the same resistance over the entire remaining region but would possess a higher resistance value in that locally blocked region due to an increased vaporization of the electrolyte solvent which results in only a slight and readily permissible temperature increase (if at all) at the blocked region. After removal of the blockage from a region of the heating element, the temperature will decrease and the vaporized solvent will cool and be reabsorbed in the electrolyte sheet. This reabsorption will result in a decrease in resistance in the region and consequently an increase in current flow.

In this way the resistance value of the heating element adjusts itself from place to place according to the temperature prevailing at each particular place. Therefore, overheating and hence burning or other damage to the heating element is completely eliminated because in every region of the heating element the resistance value automatically adjusts itself so that current consumption and hence the heat produced at that particular region does not permit the temperature thereof to increase beyond a desired maximum value. Thus, the heating element maintains a regulated temperature regardless of change in surrounding conditions.

The same positive temperature coefficient can also be achieved by using non-aqueous solvents which have a sharply decreasing dielectric constant with increasing temperature and which will over-compensate for the increase in electrical conductivity due to a sharp increase in ion mobility as the temperature increases. As the dielectric constant decreases, the impedance of the solvent to the flow of curernt increases. Hence, a sharp decrease in electric current flow can be achieved with increasing temperature by using an appropriate solvent. This foregoing characteristic of the solvent must over-compensate for the increase in electrical conductivity of the electrolyte which occurs upon heating of the electrolyte and produces an overall decrease in the electrical current flow with increasing temperature in the heating element, i.e., a positive temperature coefficient. If the solvent does not have a sharply decreasing dielectric constant with increasing temperature, over-compensation will not be achieved and hence a negative temperature coefficient condition would occur.

As a result of the selection of a solvent having the desired dielectric constant properties which will produce a positive temperature coefficient, a self-regulating optically transparent heating element is provided.

Although the present invention has been described in conjunction with preferred embodiments, it is to be understood that modifications and variations may be resorted to without departing from the spirit and scope thereof, as those skilled in the art will readily understand, such variations and modifications are considered to be within the purview and scope of the appended claims.

What is claimed is:

1. An optically transparent electrical heating element comprising (1) insulating means having a pair of relatively flat side wall portions joined together, at least one dimension of said relatively flat side wall portion being smaller than the corresponding dimension of the other relatively flat side wall portion and (2) an electrolyte encased and sealed within said insulating means, said insulating means being substantially stable toward said electrolyte and said insulating means having opposed spaced electrically conductive means on the inside face thereof in contact with said electrolyte, a portion of said electrically conductive means extending outside of said sealed portion of said insulating means for connecting said heating element with an electric current source.

2. A heating element according to claim 1, wherein each of said side wall portions comprises a polymeric sheet material.

3. A heating element according to claim 1, wherein said side wall portions are sealed to each other to provide a substantially vapor-tight casing.

4. A heating element according to claim 1, wherein said side wall portions are substantially flexible.

5. A heating element according to claim 1 characteized by a positive temperature coefficient.

6. An optically transparent heating element having a positive temperature coefficient comprising (1) a polymeric support material, (2) an electrolyte solution absorbed on said support material, and (3) insulating means substantially impervious to air and solvent vapors, substantially stable toward said electrolyte solution and encasing said electrolyte support material, said support material and insulating means each having opposed spaced conductive means in electrical contact relationship, a portion of said conductive means of said insulating material extending outside of the encased portion of said heating element for connection to an electric current source said heating element during the passage of electric current therethrough automatically increasing and decreasing the resistance of individual areas thereof in accordance with the dissipation of heat from said individual areas.

7. A heating element according to claim 6 wherein said support material and insulating means are optically transparent to render said heating element transparent.

8. A heating element according to claim 6, wherein said insulating means includes a pair of relatively flat side wall portions joined together, said electrolyte support material being encased within said side wall portions of said heating element.

9. A heating element according to claim 8, wherein at least one dimension of one of said relatively flat side wall portions is smaller than the corresponding dimension of the other relatively flat side wall portion.

10. A heating element according to claim 9, wherein said electrolyte support material is a relatively flat sheet lying between said side wall portions.

11. A heating element according to claim 10, wherein said electrolyte support material is a polymeric material

12. A heating element according to claim 11 wherein said polymeric material is a cross-linked polymer.

13. A heating element according to claim 9, wherein said insulating means comprises a flexible polymeric material which will not absorb said electrolyte.

14. A heating element according to claim 9, wherein said insulating means comprises a material that is heat-sealable.

15. A heating element according to claim 9, wherein said electrically conductive means comprises spaced electrodes at least a portion of the respective electrodes of said insulating means and of said electrolyte support material overlying each other, and said electrodes of said insulating means being adapted to be connected to respective terminals of an electrical current source.

16. A heating element according to claim 6 wherein said polymeric support material is a cellulose or cellulose derivative.

17. A heating element according to claim 6 wherein said electrolyte solution is a non-aqueous solution.

18. An electrolyte foil according to claim 17 wherein said electrolyte solution contains a dissociation organic or inorganic acid.

References Cited

UNITED STATES PATENTS

2,389,360	11/1945	Guyer et al.	219—547 X
2,441,831	5/1948	Moore	219—547
2,543,363	2/1951	Glendinning et al.	219—203
2,739,083	3/1956	Brown et al.	117—211
2,879,367	3/1959	MeLean	219—385
3,020,376	2/1962	Hofmann et al.	219—543 X
3,177,345	4/1965	Plumat	219—543
3,296,641	1/1967	Eisler	219—385
3,419,702	12/1968	Piel	338—114 X

VOLODYMYR Y. MAYEWSKY, Primary Examiner

U.S.Cl. X.R.

219—203, 286, 295, 543, 549; 338—212