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(54) Title: COLORANT-CONTAINING ELECTRORHEOLOGICAL MATERIALS

(57) Abstract

An electrorheological material containing a carrier fluid, an electrorheologically active particle and a colorant additive which can be classified as a dye, an insoluble pigment, an A-type soluble pigment, or a B-type soluble pigment. The incorporation of these colorant additives in accordance with the invention does not dramatically affect the viscosity, dynamic yield stress, static yield stress, current density or response time exhibited by the electrorheological material. The colorant-containing electrorheological material is useful for creating aesthetic visual effects in a variety of applications.

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Description

COLORANT-CONTAINING ELECTRORHEOLOGICAL MATERIALS

Technical Field

The present invention relates to fluid compositions that exhibit substantial changes in rheological properties when exposed to electric fields. More specifically, the present invention relates to electrorheological materials containing a colorant additive.

Background Art

Electrorheological materials are fluid compositions that exhibit substantial changes in rheological properties in the presence of an electric field. Electrorheological materials typically consist of (1) a carrier fluid, (2) a particle component, (3) an activator, and (4) a surfactant. The surfactant of the electrorheological material is utilized to disperse the particle component within the carrier fluid while the activator is utilized to impart electroactivity to the particle component. In the presence of an electric field, the particle component becomes organized so as to increase the apparent viscosity or flow resistance of the overall material. Therefore, by manipulating the electric field, one can selectively change the apparent viscosity or flow resistance of an electrorheological material to achieve desired results in various known devices and applications.

U.S. Patent No. 5,075,021 discloses that the observed electrorheological activity of a given material is directly dependent on the polarizability of the particle in the fluid medium. The polarizability, β, of a particle in a fluid medium is given by:

$$\beta = [(\varepsilon_2-\varepsilon_1)/(\varepsilon_2+2\varepsilon_1)]$$

where ε_1 and ε_2 are the complex permittivities of the carrier fluid and particle, respectively. In order for a given material to polarize and respond as an electrorheological material, the particle and the carrier

fluid of that material must have different complex permittivities so that $\varepsilon_2 - \varepsilon_1 \neq 0$. By definition, the complex permittivity of a fluid or particle is dependent upon both the dielectric constant and conductivity of the fluid or particle.

Over the years, many different types of electrorheological materials have been developed that are based on numerous types of particle components. These previously developed electrorheological materials may be utilized in various devices, including dampers designed for controlling vibration of a system in either an on/off or continuously variable manner. In many instances, an electrorheological material can be selected in order to provide specific performance characteristics in the particular device or application selected. For example, in a device where it is necessary or desirable to see the inner workings of a device with the human eye, an optically transparent electrorheological material will be selected such as that disclosed in U.S. Patent No. 5,075,021.

In certain other devices or applications where the electrorheological material is readily visible to an outside observer, it may be desirable to utilize an electrorheological material that has aesthetic shades of color. In fact, several electrorheological materials have been previously mentioned in the patent literature that utilize dye pigments as electrorheologically active particles. For example, U.S. Patent No. 3,484,162 describes an electroviscous recording device wherein an electrorheological material is used to create images on a substrate by releasing the material onto the substrate in response to radiation from a lamp that energizes a photoconductive cell to short circuit an electric field that maintains the material in a viscous state. Since the electrorheological materials utilized in the device must have a dark or readily visible color, carbon black or another suitable dye pigment is employed as the particle component in the material. U.S. Patent No. 3,484,162 discloses, as a specific example, a red electroviscous material containing Acetamine Rubine B and paraffin oil.

U.S. Patent No. 3,553,708 discloses another electroviscous recording device wherein materials that exhibit changes in electroactivity in the presence of actinic radiation or light and an electric field are utilized in the electrorheological material. A constant electric field is applied to the electrorheological material and a lamp is utilized to release the electrorheological material at selected intervals in response to changes in radiation. The electrorheological materials utilize various dye pigments as active particles including phthalocyanine-type compounds such as copper phthalocyanine. The carrier liquid of the materials may be any dielectric liquid such as mineral oil, chlorinated hydrocarbons, fluorinated hydrocarbons, etc.

The utilization of an ionic dye compound as the active particle component in electrorheological materials is disclosed in co-pending U.S. Patent Application Serial No. 07/806,981 entitled "Ionic Dye-Based Electrorheological Materials." The ionic dye compound preferably contains at least one aromatic nucleus that has been sulfonated and/or carboxylated to form a negatively charged system which has, in turn, been offset with a positive ion. The ionic dye-containing electrorheological material does not require the presence of actinic radiation, responds quickly to an electric field, and is useful for creating aesthetic visual effects.

Devices utilizing electrorheological materials typically have certain minimum performance requirements for the electrorheological material contained within the device. In other words, the complex permittivities of the particle and carrier fluid must be selected such that the particle will exhibit a certain minimum polarizability as defined above. In applications where it is desirable to utilize an electrorheological material having a specific color or hue, traditional electrorheological materials having dye particle components such as those described above oftentimes do not meet the minimum performance requirements of a particular device.

A need therefore exists for an electrorheological material that can not only meet the minimum performance standards of most electrorheological material devices but can also exhibit a desired color or hue for a given application.

Disclosure of Invention

The present invention is an electrorheological material that meets the minimum performance requirements of current electrorheological devices while simultaneously being useful in applications where it is desired to produce an aesthetic visual effect. The electrorheological material of the present invention comprises a carrier fluid component, a particle component and a colorant additive. The colorant additives of the invention can be selected from the group consisting of dyes, insoluble pigments, A-type soluble pigments and B-type soluble pigments, all of which will be described in more detail hereinafter. In order to minimize the impact of the colorant additive on the performance of the electrorheological material, the colorant additive of the invention is preferably utilized in an amount not to exceed about 5 percent by weight of the total material.

It has also been found that different types of colorants have correspondingly different effects on the properties of an electrorheological material and that each type of colorant can be utilized in an amount falling within a respective unique range in order to minimize or eliminate any negative impact of the colorant on the performance of the overall electrorheological material. If utilized in these respective ranges, the colorant will have essentially no detrimental effect on electrorheological material properties such as viscosity, dynamic yield stress, static yield stress, current density and response time.

The present electrorheological material may optionally contain an activator such as water or other molecule-containing hydroxyl, carboxyl or amine functionality and may also optionally contain a surfactant. The colorant additives of the invention can be utilized in combination with any electrorheological particles and are capable of providing aesthetic hues or colors that are useful in applications wherein the electrorheological material is visible to an outside observer.

Best Mode for Carrying Out the Invention

The present invention relates to an electrorheological material comprising a carrier fluid component, a particle component and a colorant additive.

The carrier fluid of the invention is a continuous liquid phase and may be selected from any of a large number of electrically insulating, hydrophobic liquids known for use in electrorheological materials. Typical liquids useful in the present invention include mineral oils, white oils, paraffin oils, chlorinated hydrocarbons such as 1-chlorotetradecane, silicone oils, transformer oils, halogenated aromatic liquids, halogenated paraffins, polyoxyalkylenes, perfluorinated polyethers, fluorinated hydrocarbons and mixtures thereof. Perfluorinated polyethers, silicone and hydrocarbon oils having viscosities of between about 0.65 and 1000 milli Pascal seconds (mPa·s) are the preferred carrier fluids of the invention. As known to those familiar with such compounds, transformer oils refer to those liquids having characteristic properties of both electrical and thermal insulation. Naturally occurring transformer oils include refined mineral oils that have low viscosity and high chemical stability. Synthetic transformer oils generally comprise chlorinated aromatics (chlorinated biphenyls and trichlorobenzene), which are known collectively as "askarels", silicone oils, and esteric liquids such as dibutyl sebacates. The carrier fluid is utilized in an amount ranging from about 50 to 95, preferably from about 60 to 85, percent by volume of the final electrorheological material. This corresponds to approximately 67 to 97, preferably 75 to 92, percent by weight when the carrier fluid and particle of the electrorheological material have a specific gravity of about 1.0 and 2.0, respectively.

The particle component can essentially be any solid which is known to exhibit electrorheological activity. Typical particle components useful in the present invention include amorphous silicas, synthetic silicas, precipitated silicas, fumed silicas, silicates, aluminum silicates, ion exchange resins and other inorganic particles such as those composed of titanium dioxide, barium titanate,

lithium hydrazinium sulfate and insulated metallic particulates. Other typical particle components useful in the present invention include polyvinyl alcohols, polyhydric alcohols, silicone ionomer reaction products, monosaccharides, porphin systems, metallo-porphin systems, poly(acene-quinone) polymers, polymeric Schiff bases, anionic surfactants, polyelectrolytes, carbonaceous particulates, and other organic and polymeric particles such as those composed of polymethacrylic acid salts and copolymers of phenol, aldehydes, olefins, ethers and/or acids. The diameter of the particles utilized herein can range from about 0.1 to 500 μm and preferably from about 1.0 to 50 μm. The particle component typically comprises from about 5 to 50, preferably from about 15 to 40, percent by volume of the total composition depending on the desired electroactivity and viscosity of the overall material. This corresponds to approximately 3 to 33, preferably 8 to 25, percent by weight when the carrier fluid and particle of the electrorheological material have a specific gravity of about 1.0 and 2.0, respectively.

The electrorheological material of the present invention may contain a small amount of an activator in combination with the particle component. Typical activators for optional use in the present invention include water and other molecules containing hydroxyl, carboxyl or amine functionality. Typical activators other than water include methyl, ethyl, propyl, isopropyl, butyl and hexyl alcohols, ethylene glycol, diethylene glycol, propylene glycol, glycerol; formic, acetic, sulfuric and lactic acids; aliphatic, aromatic and heterocyclic amines, including primary, secondary and tertiary amino alcohols and amino esters that have from 1-16 atoms of carbon in the molecule; methyl, butyl, octyl, dodecyl, hexadecyl, diethyl, diisopropyl and dibutyl amines, ethanolamine, propanolamine, ethoxyethylamine, dioctylamine, triethylamine, trimethylamine, tributylamine, ethylene-diamine, propylene-diamine, triethanolamine, triethylenetetramine, pyridine, morpholine and imidazole; and mixtures thereof. Water is the preferred activator for optional use in the present invention. When employed, the activator is utilized in an amount from about 0.1 to 10, preferably from about 0.5 to 5.0, percent by weight relative to the weight of the particle component.

A surfactant to disperse the particle component may also be utilized in the present invention. Such surfactants include known surfactants or dispersing agents such as glycerol monooleate, sorbitan sesquioleate, stearates, laurates, fatty acids, fatty alcohols, and the other surface active agents discussed in U.S. Patent No. 3,047,507 (incorporated herein by reference) but preferably comprise non-ionic surfactants such as the steric stabilizing amino-functional, hydroxy-functional, acetoxy-functional, or alkoxy-functional polysiloxanes such as those disclosed in U.S. Patent No. 4,645,614 (incorporated herein by reference). Other steric stabilizers such as graft and block copolymers may be utilized as a surfactant for the present invention and such other steric stabilizers as, for example, block copolymers of poly(ethylene oxide) and poly(propylene oxide) are disclosed in detail in U.S. Patent No. 4,772,407 (incorporated herein by reference) and in Napper, "Polymeric Stabilization of Colloidal Dispersions," Academic Press, London, 1983 (incorporated herein by reference). Still other steric stabilizers include hyperdispersants, such as HYPERMER® (ICI Americas, Inc.) and SOLSPERSE® (ICI Americas, Inc.) hyperdispersants, fluoroaliphatic polymeric esters, such as FC-430 (3M Corporation), and coupling agents including titanate, aluminate or zirconate coupling agents, such as KEN-REACT® (Kenrich Petrochemicals, Inc.) coupling agents.

The surfactant, if utilized, is preferably an amino-functional polydimethylsiloxane, a fluoroaliphatic polymeric ester, a hyperdispersant or a coupling agent. The optional surfactant may be employed in an amount ranging from about 0.1 to 20 percent by weight relative to the weight of the particle component.

The compounds that function as the additives providing color to the present electrorheological material can essentially be any known colorant. More specifically, the colorant additive can be any dye or pigment that imparts color to the electrorheological material. The distinction between a dye and a pigment for the present invention is based upon the solubility of the colorant in the carrier fluid of the electrorheological material. A colorant additive to the electro-rheological material of the present invention is identified as a dye

when it is soluble in the carrier fluid and a pigment when it is not soluble in the carrier fluid.

Since the electrorheological materials of the present invention may optionally contain an activator in combination with the particle component, a pigment must be further classified depending upon its solubility in this activator. A pigment that is soluble in the activator is identified as a soluble pigment, while a pigment that is insoluble in the activator is identified as an insoluble pigment. If an activator is not present in the electrorheological material, all pigments are considered to be insoluble pigments. For purposes of the present invention, the classification of a particular colorant as a dye, a soluble pigment or an insoluble pigment is therefore dependent on the type of electrorheological material with which the colorant is utilized.

It has been found that the soluble pigments of the invention can interact with the surface of the particle so as to affect the particle's overall conductivity. It has further been found that a soluble pigment affects the overall electrorheological activity of the material in different ways depending on how the soluble pigment affects the conductivity of the particle. For purposes of this invention, "A-type soluble pigment" herein refers to a soluble pigment which increases the conductivity of the particle component, while "B-type soluble pigment" herein refers to a soluble pigment which decreases the conductivity of the particle component.

The colorant additive of the present invention can be selected from any of the well-known classes of organic colorants including phthalocyanine, metal complex, nitroso, nitro, monoazo, disazo, trisazo, polyazo, azoic, stilbene, diphenylmethane, triarylmethane, xanthene, acridine, quinoline, methine, thiazole, indamine, azine, oxazine, thiazine, sulphur, lactone, anthroquinone and indigoid classes. A detailed description of colorants from these classes can be found in E. N. Abrahart's Dyes and Their Intermediates, Chemical Publishing, New York, 1977 (hereinafter referred to as Abrahart), the entire disclosure of which is incorporated herein by reference. The colorant additive of the present invention also can be selected from any

inorganic powders well known to those skilled in the art of paint and ink manufacturing.

Examples of colorant additives suitable for use in the present invention are given below. The colorants are listed with their commercial name followed by the Colour Index (CI) number in parenthesis. The Colour Index system was set up under the joint sponsorship of the American Association of Textile Chemists and Colorists (AATCC) in the United States and the Society of Dyers and Colourists in Great Britain. Volumes I, II, III, and IV of the well-known Colour Index are incorporated herein by reference. The complete structures of the following compounds can be found either in Abrahart (pp. 92-107), in the 1990-1991 Aldrich Catalog Handbook of Fine Chemicals, Aldrich Chemical Company, Inc., Milwaukee, 1990, or in W. M. Morgans's Outlines of Paint Technology, Halsted Press, New York, 1990, all of which are incorporated herein by reference.

Examples of ionic organic colorants useful in the present invention include Naphthol Green B (10020), Naphthol Yellow S (10316), Orange II (15510), Mordant Yellow 10 (14010), Brilliant Crocein MOO (27290), Fast Blue BB salt (37155), Brilliant Green (42040), Trisodium salt of Aurintricarboxylic acid (43810), Eosin Y (45380), Basic Yellow 11 (48055), Primulin (49000), Acid Green 25 (61570), Indigo Carmine (73015), Tetrasodium salt of Copper Phthalocyanine-tetrasulfonic acid (74160-Copper Phthalocyanine), Acid Red 74 (13355), Acid Blue 135 (13385), Acid Red 14 (14720), Mordant Black 11 (14645), Mordant Black 1 (15710), Acid Red 25 (16050), Acid Orange 10 (16230), Direct Red 70 (16081), Acid Red 29 (16570), Acid Black 31 (17580), Acid Blue 6 (17185), Mordant Black 38 (18160), Direct Orange 75 (17840), Direct Red 65 (17870), Acid Yellow 17 (18965), Acid Yellow 23 (19140), Mordant Red 7 (18760), Mordant Orange 26 (19325), Acid Red 214 (19355), Direct Orange 18 (20215), Orange II (15510), Acid Black 17 (20350), Direct Brown 2, (22311), Direct Blue 1 (24410), Acid Red 99 (23285), Acid Yellow 42 (22910), Direct Yellow 12 (24895), Direct Red 81 (28160), Acid Red 148 (26665), Direct Yellow 49 (29035), Direct Red 23 (29160), Direct Black 38 (30235), Direct Brown 54 (31735), Direct Blue 71 (34140), Direct Blue 78 (34200), Direct Red 80 (35780), Naphthol

Yellow S (10316), Martius Yellow Sodium Salt (10315), Acridine Orange (46005), Janus Green B (11050), Chrysoidin (11270), Alcian Yellow (12840), Alizarin Yellow GG (14025), Nitrazine Yellow (14890), Crocein Orange G (15970), Flavazin L (18820), Thiazol Yellow G (19540), Naphthol Blue Black (20470), Bismark Brown Y (21000), Acid Red 97 (22890), Evans Blue (23860), Direct Red 75 (25380), Ponceau S (27195), Calcomine Orange 2RS (29156), Diazo Red RC (37120), Fluorescent Brightener 28 (40622), Fast Blue RR Salt (37155), Patent Blue VF (42045), Basic Fuchsin (42510), Victoria Blue R (44040), Rhodamine B (45170), Rose Bengal (45440), Astrazon Orange G (48035), Neutral Red (50040), Celestine Blue (51050), Toluidine Blue O (52040), Alizaarin Red S monohydrate (58005), Reactive Blue 2 (61211), Acid Black 48 (65005), and Reactive Blue 15 (74459).

Examples of non-ionic organic colorants useful in the present invention include Disperse Yellow 9 (10375), Disperse Orange 3 (11005), Methyl Yellow (11020), N,N-Dimethyl-4,4'-azodianiline (11025), Disperse Orange 1 (11080), Disperse Red 1 (11110), Disperse Red 13 (11115), Fast Garnet GBC Base (11160), Fat Brown RR (11285), Mordant Brown 48 (11300), Mordant Brown 4 (11335), 4-Phenylazophenol (11800), Disperse Yellow 3 (11855), Mordant Brown 6 (11875), Mordant Brown 24 (11880), Sudan Orange G (11920), Fat Brown B (12010), Sudan I (12055), Orange OT (12100), Toluidine Red (12120), Sudan II (12140), Methyl Red (13020), Malachite Green Base (42000B), Pararosaniline Base (42500), Rosolic Acid (43800), Fluorescein (45350.1), 4',5'-Dibromofluorescein (45370.1), Eosin Y Free Acid (45380.2), Diiodofluorescein (45425.1), Acridine Orange Base (46005), Quinoline Yellow Spirit Soluble (47000), Indophenol Blue (49700), Bismarck Brown R (21010), Sudan Red 7B (26050), Disperse Orange 13 (26080), Disperse Yellow 7 (26090), Sudan III (26100), Sudan IV (26105), Sudan Red B (26110), Oil Red EGN (26120), Oil Red 0 (26125), Sudan Black B (26150), Direct Yellow 62 (36900), 2-Methyl-4-nitroaniline (37100), Fast Red ITR (37150), Fast Blue RR (37155), Fast Violet B (37165), Fast Blue BB (37175), Naphthol AS (37505), Mordant Orange 1 (14030), Nigrosin Alcohol Soluble (50415), Methylene Violet Bernthsen (52041), Alizarin (58000), Quinizarin (58050), Purpurin (58205), Quinalizarin (58500), Disperse Orange 11 (60700), Basic Blue 47 (61111), Disperse Blue 14

(61500), Disperse Blue 3 (61505), Solvent Blue 35 (61554), Oil Blue N (61555), Solvent Green 3 (61565), Indigo (73000), Copper Phthalocyanine (74160), Hematoxylin (75290), Carminic Acid (75470), 2-Hydroxy-1,4-naphthoquinone (75480), Lapachol (75490), and 5-Chloroisatoic Anhydride (5-CIA).

Examples of inorganic colorants useful in the present invention include Titanium Dioxide (CI pigment white 6), Zinc Oxide (CI pigment white 4), Antimony Oxide (CI pigment white 11), Lead Chromes, (CI pigment yellow 34, pigment red 104), Zinc Chromes (CI pigment yellow 36 and 36.1), Cadmium Colours (CI pigment yellow 37, pigment red 108), Ferrite Yellow (CI pigment yellow 42), Iron Red Oxide (CI pigment red 101), Red Lead (CI pigment red 105), Ultramarine Blue (CI pigment blue 29), Prussian Blues (CI pigment blue 27), Cobalt Blue (CI pigment blue 28), Chrome Greens (CI pigment green 15), Chromium Oxide (CI pigment green 17), Viridian (CI pigment green 18) and Carbon Black (CI pigment black 7).

The colorant additive of the present invention can also be an ionic dye compound as defined in co-pending U.S. Patent Application Serial No. 07/806,981, entitled "Ionic Dye-Based Electrorheological Materials," which is incorporated herein by reference.

The selection of a particular colorant additive for utilization in the electrorheological material of the present invention is based upon the hue, tone and depth of colors desired for the intended application.

As stated above, the colorant additive of the present invention is preferably utilized in an amount less than about 5 percent by weight of the total electrorheological material. It has been found that the presence of a colorant additive in excess of about 5 percent by weight can, in some cases, interfere with the electrorheological activity of the overall material. Specifically, it has been found that the preferred range of amounts of dyes and insoluble pigments for use in the invention is from about 0.01 to 5.0, preferably from about 0.1 to 3.0, percent by weight of the total material. Furthermore, it has been determined that an A-type soluble pigment is preferably utilized in an amount ranging from about 0.01 to 3.0, preferably about 0.1 to 1.0,

percent by weight of the total electrorheological material, while a B-type soluble pigment is preferably utilized in an amount ranging from about 0.01 to 0.50, preferably from about 0.01 to 0.10, percent by weight of the total electrorheological material. In applications where it is desirable for the performance of the electrorheological material to mimic the performance of the corresponding non-colorant-containing electrorheological material, the above preferred ranges should be strictly adhered to. However, if a slight compromise in performance can be tolerated, the colorant can be utilized in amounts outside of the preferred ranges.

Since the solubilities of colorants in various materials are not well known, a slight degree of experimentation will be required to classify any given colorant in accordance with the present invention. In order to classify a colorant as a dye or pigment, the colorant may be added to a sample of the particular carrier fluid selected for the electrorheological material. If the colorant is soluble in the carrier fluid, the colorant would be classified as a dye, while insolubility would result in a classification of the colorant as a pigment. To further classify a pigment as an insoluble or soluble pigment, the pigment would be added to a sample of the activator selected, if any, for the electrorheological material so as to determine the solubility of the pigment in the activator. To determine whether a soluble pigment is an A-type or B-type soluble pigment, the conductivity of the suspended particles in the electrorheological material are determined from A.C. impedance measurements which are described in more detail hereinafter.

The electrorheological materials of the present invention can be prepared by simply mixing together the carrier fluid, the particle component, the colorant additive and optionally any activator and/or surfactant. The colorant additive can be used in either a "substantially anhydrous" form obtained through the removal of moisture adsorbed on the surface of the dye or pigment by methods known to those skilled in the art or in an "as received" condition, which will inherently contain physically adsorbed moisture. The ingredients of the electrorheological materials may be initially mixed

together by hand with a spatula or the like and then subsequently more thoroughly mixed with a mechanical mixer or shaker, or dispersed with an appropriate milling device such as a ball mill, sand mill, attritor mill, paint mill, etc., in order to create smaller particles and a more stable suspension. It is also possible to incorporate the colorant additive into an existing formulated electrorheological material through the use of the mixing techniques described above.

Evaluation of the mechanical and electrical properties of the electrorheological materials of the present invention, as well as other electrorheological materials, can be carried out through the use of concentric cylinder couette rheometry. The theory which provides the basis for this technique is adequately described by S. Oka in Rheology, Theory and Applications , volume 3, F. R. Eirich, ed., Academic Press: New York, 1960, pages 17-82, which is incorporated herein by reference. The information that can be obtained from a concentric cylinder rheometer includes data relating mechanical shear stress to shear strain, the static yield stress and the electrical current density as a function of shear rate. For electrorheological materials, the shear stress versus shear rate data can be modeled after a Bingham plastic in order to determine the dynamic yield stress and viscosity. The test geometry that is utilized by these rheometers for the characterization of ER materials is a simple concentric cylinder couette cell configuration. The material is placed in the annulus formed between an inner cylinder of radius R_1 and an outer cylinder of radius R_2. One of the cylinders is then rotated with an angular velocity Ω while the other cylinder is held motionless. The relationship between the shear stress and the shear strain rate is then derived from this angular velocity and the torque, T, applied to maintain or resist it.

A method that can be used to determine the response times associated with electrorheological materials of the present invention is described in detail in a paper by Weiss and Carlson presented on October 16, 1991 at the 3rd International Conference on ER Fluids (Carbondale, Illinois), the entire disclosure of which is incorporated herein by reference. This technique, which is based upon dielectric or

impedance spectroscopy, allows for the measurement of the time necessary for individual particles to respond to a small change in electric field. In this technique, data describing the electrical response of a material is obtained through the application of a small alternating current signal to the sample over a broad frequency range. Analysis of this data using circuit theory provides a description of the material's dielectric properties. It is known that electrorheological materials undergo a Debye-like relaxation process. According to Debye theory the time dependent dielectric data obtained for an electrorheological material using impedance spectroscopy can be used to determine the relaxation time associated with the polarization decay process. This relaxation time for an electrorheological material can be obtained either at the angular frequencies where maxima are observed in the dielectric loss and/or loss tangent data or through the use of Cole-Cole arc diagrams. This measurement provides a lower limit for the time interval in which an electrorheological material response can be expected. In other words, the electrorheological material can never exhibit a response to an electric field faster than the relaxation time indicated by this measurement.

The conductivity of the suspended particles utilized in the electrorheological material of the invention can be determined from the previously described A.C. impedance measurements. The well known Maxwell-Wagner theory of heterogeneous dielectrics can be utilized to determine the particle conductivity from the complex dielectric data. A more detailed description of this theory is provided by J. B. Hasted in Aqueous Dielectrics, Chapman & Hall Publishers, London, 1989, the entire contents of which are herein incorporated by reference.

The following examples are given to illustrate the invention and should not be construed to limit the scope of the invention.

Example 1 - Preparation of Base Material

An electrorheological material is prepared through the addition of 1000.0 g light mineral oil (Johnson Matthey, Alpha Products), 150.0 g precipitated silica (Union Carbide Co.), 4.50 g

distilled water and 30.0 g glycerol monooleate (Witco Chemical Co.). The properties of this electrorheological material are evaluated using concentric cylinder couette rheometry and impedance spectroscopy. This electrorheological material comprises the base material to which all colorants are added and tested for purposes of the following examples.

Example 2 - Classification of Colorants

Approximately 5.00 x 10^{-3} g of four different organic colorants, namely Acridine Orange Salt (46005), Oil Red O (26125), Copper Phthalocyanine (74160) and Acid Green 25 Salt (61570) obtained from Aldrich Chemical Co., are added to separate 5.00 g samples of the light mineral oil and distilled water of Example 1. The solubility of Oil Red O in the mineral oil sample is evident through the appearance of mixing lines and the disappearance of all solid particulates. In the case of Copper Phthalocyanine, Acid Green 25 Salt and Acridine Orange Salt, the presence of insoluble particulates is observed in the mineral oil samples. Both the Acridine Orange Salt and Acid Green 25 Salt colorants are observed to dissolve in the water samples. However, both the Oil Red O and Copper Phthalocyanine colorants are observed to be insoluble in the water samples. This example demonstrates that for utilization as an additive to the electrorheological base material prepared in Example 1, these four colorants can be classified as follows: Acridine Orange Salt (soluble pigment); Oil Red O (dye); Acid Green 25 Salt (soluble pigment); and Copper Phthalocyanine (insoluble pigment).

Examples 3-6 - Determination of A-Type v. B-Type Solubility

For Example 3, a total of 46.64 g of the electrorheological base material prepared in Example 1 is placed into a sample vessel and a total of 4.67 x 10^{-3} g Acid Green 25 Salt is added. This amount of the insoluble pigment is equivalent to about 0.01 percent by weight of the total electrorheological material. The insoluble pigment is adequately dispersed in the base material through the use of a high speed mechanical disperser.

In Examples 4-6, the same procedure as described for Example 3 is used for the addition of various amounts of Acid Green 25 Salt or Acridine Orange Salt to a 46.64 g sample of the electrorheological base material prepared in Example 1. The weight amount and type of the soluble pigment this is used in each Example is described below.

	Soluble Pigment	Weight Amount of Pigment Added (grams)	Weight Percent of Total Material (%)
Example 4	Acid Green 25 Salt	4.67×10^{-2}	0.10
Example 5	Acridine Orange Salt	4.67×10^{-3}	0.01
Example 6	Acridine Orange Salt	4.67×10^{-2}	0.10

The dielectric data for the electrorheological base material of Example 1 and the soluble pigment-containing electrorheological materials of Examples 3-6 are obtained using impedance spectroscopy. The conductivity of the suspended particles present in the electrorheological materials are determined from these measurements using Maxwell-Wagner theory as previously defined. The measurements obtained for Examples 1 and 3-6 are shown below in Table 1.

Table 1

Acid Green 25 Salt	Example 1	Example 3	Example 4
Weight % of soluble pigment added	0.00	0.01	0.10
Particle conductivity (μs/m)	0.0042	0.0055	0.0096

Acridine Orange Salt	Example 1	Example 5	Example 6
Weight % of soluble pigment added	0.00	0.01	0.10
Particle conductivity (μs/m)	0.0042	0.0040	0.0022

Examples 3-4 demonstrate that Acid Green 25 Salt is a soluble pigment that increases the conductivity of the particle component. Examples 5-6 demonstrate that Acridine Orange Salt is a soluble pigment that decreases the conductivity of the particle component.

Therefore, for utilization as a soluble pigment additive to the electrorheological base material in Example 1, Acid Green 25 Salt can be classified as an A-type soluble pigment while Acridine Orange Salt can be classified as a B-type soluble pigment.

Examples 7-12

In Examples 7-12, the same procedure as described for Example 3 is used for the addition of various amounts of copper phthalocyanine to a 46.64 g sample of the electrorheological base material prepared in Example 1. The weight amount of the insoluble pigment that is used in each Example is described below.

	Weight Amount of Pigment Added (grams)	Weight Percent of Total Material (%)
Example 7	4.67×10^{-3}	0.01
Example 8	4.67×10^{-2}	0.10
Example 9	2.33×10^{-1}	0.50
Example 10	4.66×10^{-1}	1.00
Example 11	1.40	3.00
Example 12	2.33	5.00

The mechanical and electrical properties of the colorant containing electrorheological materials of Examples 7-12 are evaluated through the use of concentric cylinder couette rheometry and impedance spectroscopy. The measurements obtained for Examples 7-12 are shown in Table 2. The test data obtained for the

electrorheological base material (Example 1) also is shown in this Table for comparison.

Table 2

Description of the Measured Property	Ex. #1	Ex. #7	Ex. #8	Ex. #9	Ex. #10	Ex. #11	Ex. #12
Viscosity (mPa-s)	160	142	147	180	181	221	196
Zero-Field Yield Stress (Pa)	11	13	8	11	13	24	18
Dynamic Yield Stress (Pa) at 3.0 kV/mm	291	294	295	304	324	287	304
Static Yield Stress (Pa) at 3.0 kV/mm	325	323	329	324	348	312	344
Current Density ($\mu A/cm^2$) at 3.0 kV/mm	0.82	1.03	1.33	0.31	0.87	1.02	0.99
Relaxation Time (ms)	22.6	15.6	15.6	12.7	12.7	13.3	16.8

Examples 7-12 demonstrate that an insoluble pigment can be used as a colorant to an electrorheological material in amounts up to about 5.0 percent by weight with respect to the total material without any detrimental effect on the properties exhibited by the electrorheological material. In fact, the data shown in Table 2 indicates that a slight improvement in relaxation time and the electric field-induced yield stress (static and dynamic) is observed upon incorporation of the insoluble pigment additive.

Examples 13-18

In Examples 13-18, the same procedure as described for Example 3 is used for the addition of various amounts of Oil Red O to a 46.64 g sample of the electrorheological base material prepared in

Example 1. The weight amount of the dye that is used in each Example is described below.

	Weight Amount of Dye Added (grams)	Weight Percent of Total Material (%)
Example 13	4.67×10^{-3}	0.01
Example 14	4.67×10^{-2}	0.10
Example 15	2.33×10^{-1}	0.50
Example 16	4.66×10^{-1}	1.00
Example 17	1.40	3.00
Example 18	2.33	5.00

The mechanical and electrical properties of the colorant containing electrorheological materials of Examples 13-18 are evaluated through the use of concentric cylinder couette rheometry and impedance spectroscopy. The measurements obtained for Examples 13-18 are shown in Table 3. The test data obtained for the electrorheological base material of Example 1 also is shown in this Table for comparison.

Table 3

Description of the Measured Property	Ex. #1	Ex. #13	Ex. #14	Ex. #15	Ex. #16	Ex. #17	Ex. #18
Viscosity (mPa-s)	160	167	149	148	144	153	162
Zero-Field Yield Stress (Pa)	11	6	8	9	5	3	7
Dynamic Yield Stress (Pa) at 3.0 kV/mm	291	294	291	279	279	234	244
Static Yield Stress (Pa) at 3.0 kV/mm	325	327	345	317	295	290	300
Current Density ($\mu A/cm^2$) at 3.0 kV/mm	0.82	0.48	0.59	1.33	1.05	0.28	0.88
Relaxation Time (ms)	22.6	16.2	21.1	18.1	14.7	19.6	20.1

Examples 13-18 demonstrate that a dye can be used as a colorant to an electrorheological material in amounts up to about 5.0 percent by weight with respect to the total material without any detrimental effect on the properties exhibited by the electrorheological material. A slight decrease in the electric field-induced yield stress (static and dynamic) is observed upon incorporation of the dye additive as shown in Table 3. However, this decrease is not significant until after more than 5.0 weight percent of the colorant is added to the electrorheological material.

Examples 19-22

In Examples 19-22, the same procedure as described for Example 3 is used for the addition of various amounts of Acid Green 25 Salt to a 46.64 g sample of the electrorheological base material prepared in Example 1. The weight amount of the A-type soluble pigment that is used in each Example is described below.

	Weight Amount of Pigment Added (grams)	Weight Percent of Total Material (%)
Example 19	2.33×10^{-1}	0.50
Example 20	4.66×10^{-1}	1.00
Example 21	1.40	3.00
Example 22	2.33	5.00

The mechanical and electrical properties of the colorant containing electrorheological materials of Examples 3, 4, and 19-22 are evaluated through the use of concentric cylinder couette rheometry and impedance spectroscopy. The measurements obtained for Examples 3, 4, and 19-22 are shown in Table 4. The test data obtained for the electrorheological base material prepared in Example 1 also is shown in this Table for comparison.

Table 4

Description of the Measured Property	Ex. #1	Ex. #3	Ex. #4	Ex. #19	Ex. #20	Ex. #21	Ex. #22
Viscosity (mPa-s)	160	151	153	141	141	157	149
Zero-Field Yield Stress (Pa)	11	9	8	10	13	14	11
Dynamic Yield Stress (Pa) at 3.0 kV/mm	291	300	325	325	315	346	arc
Static Yield Stress (Pa) at 3.0 kV/mm	325	332	335	330	343	365	arc
Current Density ($\mu A/cm^2$) at 3.0 kV/mm	0.82	0.63	1.42	0.51	1.50	0.17	arc
Relaxation Time (ms)	22.6	17.6	10.4	11.6	11.7	12.6	17.5

Examples 3, 4, and 19-22 demonstrate an A-type soluble pigment can be utilized as a colorant to an electrorheological material in an amount up to about 3.0 percent by weight with respect to the total material without any detrimental effect on the properties exhibited by the electrorheological material. In fact, the data shown in Table 4 indicates that a slight improvement in relaxation time and the electric field-induced yield stress (static and dynamic) is observed upon incorporation of this A-type soluble pigment additive. If more than about 3.0 weight percent of this A-type soluble pigment is used as the colorant additive, inferior electrical performance is observed.

Examples 23-26

In Examples 23-26 the same procedure as described for Example 3 is used for the addition of various amounts of Acridine Orange Salt to a 46.64 g sample of the electrorheological base material prepared in Example 1. The weight amount of the B-type soluble pigment that is used in each Example is described below.

	Weight Amount of Pigment Added (grams)	Weight Percent of Total Material (%)
Example 23	2.33×10^{-1}	0.50
Example 24	4.66×10^{-1}	1.00
Example 25	1.40	3.00
Example 26	2.33	5.00

The mechanical and electrical properties of the colorant containing electrorheological materials of Examples 5, 6, and 23-26 are evaluated through the use of concentric cylinder couette rheometry and impedance spectroscopy. The measurements obtained for Examples 5, 6, and 23-26 are shown in Table 5. The test data obtained for the electrorheological base material prepared in Example 1 also is shown in this Table for comparison.

Table 5

Description of the Measured Property	Ex. #1	Ex. #5	Ex. #6	Ex. #23	Ex. #24	Ex. #25	Ex. #26
Viscosity (mPa-s)	160	161	162	149	150	175	209
Zero-Field Yield Stress (Pa)	11	5	8	1	2	1	19
Dynamic Yield Stress (Pa) at 3.0 kV/mm	291	264	252	156	100	104	173
Static Yield Stress (Pa) at 3.0 kV/mm	325	335	295	186	75	89	170
Current Density ($\mu A/cm^2$) at 3.0 kV/mm	0.82	0.71	0.48	0.64	0.51	0.80	0.52
Relaxation Time (ms)	22.6	23.7	39.8	331.0	*****	*****	*****

[***** = high dielectric loss]

As indicated by the data shown in Table 5 for Examples 5, 6, and 23-26, the incorporation of a B-type soluble pigment into an electrorheological material decreases the observed electric field-induced yield stress (static and dynamic) exhibited by the base material. In addition, the relaxation time associated with the electrorheological material is observed to increase upon the incorporation of this colorant. The data indicate that this B-type soluble pigment can be utilized as a colorant to an electrorheological material in an amount up to about 0.5 percent by weight without a significantly detrimental effect on the properties exhibited by the electrorheological material. If more than about 0.5 percent by weight of this B-type soluble pigment is utilized, undesirably high viscosities are experienced and dielectric loss occurs.

The preceding Examples 3-26 demonstrate that the incorporation of a colorant into an electrorheological material can be achieved without seriously affecting the electrorheological material.

The proper classification of a colorant as a dye, insoluble pigment, A-type soluble pigment, or B-type soluble pigment can further assist in the proper selection of the amount of colorant to be utilized so as to minimize interference with basic electrorheological performance.

Claims

What is claimed is:

1. An electrorheological material comprising a carrier fluid, a particle component, and a colorant additive.

2. An electrorheological material according to Claim 1 wherein the carrier fluid is selected from the group consisting of mineral oils, white oils, paraffin oils, chlorinated hydrocarbons such as 1-chlorotetradecane, silicone oils, transformer oils, halogenated aromatic liquids, halogenated paraffins, polyoxyalkylenes, perfluorinated polyethers, fluorinated hydrocarbons and mixtures thereof.

3. An electrorheological material according to Claim 2 wherein the carrier fluid is a perfluorinated polyether or a silicone or hydrocarbon oil having a viscosity of between about 0.65 and 1000 mPa·s.

4. An electrorheological material according to any one of Claims 1 to 3 wherein the particle component is selected from the group consisting of amorphous silicas; synthetic silicas; precipitated silicas; fumed silicas; silicates; aluminum silicates; ion exchange resins and other inorganic particles such as those composed of titanium dioxide, barium titanate, lithium hydrazinium sulfate; insulated metallic particulates; polyvinyl alcohols; polyhydric alcohols; silicone ionomer reaction products; monosaccharides; porphin systems; metallo-porphin systems; poly(acene-quinone) polymers; polymeric Schiff bases; anionic surfactants; polyelectrolytes; carbonaceous particulates; and other organic and polymeric particles such as those composed of polymethacrylic acid salts and copolymers of phenol, aldehydes, olefins, ethers and/or acids.

5. An electrorheological material according to Claim 4 wherein the diameter of the particle component is in the range from about 0.1 to 500 μm.

6. An electrorheological material according to Claim 5 wherein the diameter is in the range from about 1.0 to 50 μm.

7. An electrorheological material according to any one of the preceding claims wherein the colorant additive belongs to a class selected from the group consisting of phthalocyanine, metal complex, nitroso, nitro, monoazo, disazo, trisazo, polyazo, azoic, stilbene, diphenylmethane, triarylmethane, xanthene, acridine, quinoline, methine, thiazole, indamine, azine, oxazine, thiazine, sulphur, lactone, anthroquinone and indigoid colorant classes.

8. An electrorheological material according to any one of Claims 1 to 6 wherein the colorant additive is an ionic organic colorant selected from the group consisting of Naphthol Green B (10020), Naphthol Yellow S (10316), Orange II (15510), Mordant Yellow 10 (14010), Brilliant Crocein MOO (27290), Fast Blue BB salt (37155), Brilliant Green (42040), Trisodium salt of Aurintricarboxylic acid (43810), Eosin Y (45380), Basic Yellow 11 (48055), Primulin (49000), Acid Green 25 (61570), Indigo Carmine (73015), Tetrasodium salt of Copper Phthalocyanine-tetrasulfonic acid (74160-Copper Phthalocyanine), Acid Red 74 (13355), Acid Blue 135 (13385), Acid Red 14 (14720), Mordant Black 11 (14645), Mordant Black 1 (15710), Acid Red 25 (16050), Acid Orange 10 (16230), Direct Red 70 (16081), Acid Red 29 (16570), Acid Black 31 (17580), Acid Blue 6 (17185), Mordant Black 38 (18160), Direct Orange 75 (17840), Direct Red 65 (17870), Acid Yellow 17 (18965), Acid Yellow 23 (19140), Mordant Red 7 (18760), Mordant Orange 26 (19325), Acid Red 214 (19355), Direct Orange 18 (20215), Orange II (15510), Acid Black 17 (20350), Direct Brown 2, (22311), Direct Blue 1 (24410), Acid Red 99 (23285), Acid Yellow 42 (22910), Direct Yellow 12 (24895), Direct Red 81 (28160), Acid Red 148 (26665), Direct Yellow 49 (29035), Direct Red 23 (29160), Direct Black 38 (30235), Direct Brown 54 (31735), Direct Blue 71 (34140), Direct Blue 78 (34200), Direct Red 80 (35780), Naphthol Yellow S (10316), Martius Yellow Sodium Salt (10315), Acridine Orange (46005), Janus Green B (11050), Chrysoidin (11270), Alcian Yellow (12840), Alizarin Yellow GG (14025), Nitrazine Yellow (14890), Crocein Orange G (15970), Flavazin L (18820), Thiazol Yellow G (19540), Naphthol Blue Black (20470), Bismark Brown Y

(21000), Acid Red 97 (22890), Evans Blue (23860), Direct Red 75 (25380), Ponceau S (27195), Calcomine Orange 2RS (29156), Diazo Red RC (37120), Fluorescent Brightener 28 (40622), Fast Blue RR Salt (37155), Patent Blue VF (42045), Basic Fuchsin (42510), Victoria Blue R (44040), Rhodamine B (45170), Rose Bengal (45440), Astrazon Orange G (48035), Neutral Red (50040), Celestine Blue (51050), Toluidine Blue O (52040), Alizaarin Red S monohydrate (58005), Reactive Blue 2 (61211), Acid Black 48 (65005), and Reactive Blue 15 (74459).

9. An electrorheological material according to any one of Claims 1 to 6 wherein the colorant additive is a non-ionic organic colorant selected from the group consisting of Disperse Yellow 9 (10375), Disperse Orange 3 (11005), Methyl Yellow (11020), N,N-Dimethyl-4,4'-azodianiline (11025), Disperse Orange 1 (11080), Disperse Red 1 (11110), Disperse Red 13 (11115), Fast Garnet GBC Base (11160), Fat Brown RR (11285), Mordant Brown 48 (11300), Mordant Brown 4 (11335), 4-Phenylazophenol (11800), Disperse Yellow 3 (11855), Mordant Brown 6 (11875), Mordant Brown 24 (11880), Sudan Orange G (11920), Fat Brown B (12010), Sudan I (12055), Orange OT (12100), Toluidine Red (12120), Sudan II (12140), Methyl Red (13020), Malachite Green Base (42000B), Pararosaniline Base (42500), Rosolic Acid (43800), Fluorescein (45350.1), 4',5'-Dibromofluorescein (45370.1), Eosin Y Free Acid (45380.2), Diiodofluorescein (45425.1), Acridine Orange Base (46005), Quinoline Yellow Spirit Soluble (47000), Indophenol Blue (49700), Bismarck Brown R (21010), Sudan Red 7B (26050), Disperse Orange 13 (26080), Disperse Yellow 7 (26090), Sudan III (26100), Sudan IV (26105), Sudan Red B (26110), Oil Red EGN (26120), Oil Red 0 (26125), Sudan Black B (26150), Direct Yellow 62 (36900), 2-Methyl-4-nitroaniline (37100), Fast Red ITR (37150), Fast Blue RR (37155), Fast Violet B (37165), Fast Blue BB (37175), Naphthol AS (37505), Mordant Orange 1 (14030), Nigrosin Alcohol Soluble (50415), Methylene Violet Bernthsen (52041), Alizarin (58000), Quinizarin (58050), Purpurin (58205), Quinalizarin (58500), Disperse Orange 11 (60700), Basic Blue 47 (61111), Disperse Blue 14 (61500), Disperse Blue 3 (61505), Solvent Blue 35 (61554), Oil Blue N (61555), Solvent Green 3 (61565), Indigo (73000), Copper Phthalocyanine (74160), Hematoxylin (75290), Carminic Acid

(75470), 2-Hydroxy-1,4-naphthoquinone (75480), Lapachol (75490), and 5-Chloroisatoic Anhydride (5-CIA).

10. An electrorheological material according to any one of Claims 1 to 6 wherein the colorant additive is an inorganic colorant selected from the group consisting of Titanium Dioxide (CI pigment white 6), Zinc Oxide (CI pigment white 4), Antimony Oxide (CI pigment white 11), Lead Chromes, (CI pigment yellow 34, pigment red 104), Zinc Chromes (CI pigment yellow 36 and 36.1), Cadmium Colours (CI pigment yellow 37, pigment red 108), Ferrite Yellow (CI pigment yellow 42), Iron Red Oxide (CI pigment red 101), Red Lead (CI pigment red 105), Ultramarine Blue (CI pigment blue 29), Prussian Blues (CI pigment blue 27), Cobalt Blue (CI pigment blue 28), Chrome Greens (CI pigment green 15), Chromium Oxide (CI pigment green 17), Viridian (CI pigment green 18) and Carbon Black (CI pigment black 7).

11. An electrorheological material according to any one of the preceding claims further comprising an activator.

12. An electrorheological material according to Claim 11 wherein the activator is water and other molecules containing hydroxyl, carboxyl or amine functionality.

13. An electrorheological material according to Claim 12 wherein the activator is selected from the group consisting of water, methyl, ethyl, propyl, isopropyl, butyl and hexyl alcohols, ethylene glycol, diethylene glycol, propylene glycol, glycerol; formic, acetic, sulfuric and lactic acids; aliphatic, aromatic and heterocyclic amines, including primary, secondary and tertiary amino alcohols and amino esters that have from 1-16 atoms of carbon in the molecule; methyl, butyl, octyl, dodecyl, hexadecyl, diethyl, diisopropyl and dibutyl amines, ethanolamine, propanolamine, ethoxyethylamine, dioctylamine, triethylamine, trimethylamine, tributylamine, ethylene-diamine, propylene-diamine, triethanolamine, triethylenetetramine, pyridine, morpholine and imidazole; and mixtures thereof.

14. An electrorheological material according to Claim 13 wherein the activator is water.

15. An electrorheological material according to any one of the preceding claims further comprising a surfactant.

16. An electrorheological material according to Claim 15 wherein the surfactant is selected from the group consisting of glycerol monooleate; sorbitan sesquioleate; stearates; laurates; fatty acids; fatty alcohols; steric stabilizing amino-functional, hydroxy-functional, acetoxy-functional, or alkoxy-functional polysiloxanes; block copolymers of poly(ethylene oxide) and poly(propylene oxide); hyperdispersants; fluoroaliphatic polymeric esters; and coupling agents such as titanate, aluminate or zirconate coupling agents.

17. An electrorheological material according to Claim 16 wherein the surfactant is an amino-functional polydimethylsiloxane, a fluoro-aliphatic polymeric ester, hyperdispersant, or a coupling agent.

18. An electrorheological material as claimed in any one of the preceding claims comprising from about 50 to 95 percent by volume of the carrier fluid, from about 5 to 50 percent by volume of the particle component, and from about 0.01 to 5.0 percent by weight of the colorant additive.

19. An electrorheological material according to Claim 18 wherein the carrier fluid is present in an amount from about 60 to 85 percent by volume, and the particle component is present in an amount from about 15 to 40 percent by volume.

20. An electrorheological material according to Claim 18 wherein the colorant additive is a dye or an insoluble pigment and is present in an amount ranging from about 0.1 to 3.0 percent by weight of the electrorheological material.

21. An electrorheological material according to Claim 18 wherein the colorant additive is an A-type soluble pigment and is

present in an amount ranging from about 0.01 to 3.0 percent by weight of the electrorheological material.

22. An electrorheological material according to Claim 21 wherein the colorant additive is present in an amount ranging from about 0.1 to 1.0 percent by weight.

23. An electrorheological material according to Claim 18 wherein the colorant additive is a B-type soluble pigment and is present in an amount ranging from about 0.01 to 0.50 percent by weight of the electrorheological material.

24. An electrorheological material according to Claim 23 wherein the colorant additive is present in an amount ranging from about 0.01 to 0.10 percent by weight.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US93/01567

A. CLASSIFICATION OF SUBJECT MATTER

IPC(5) :C10M 171/00 169/04
US CL :252/73,74,572

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 252/73, 74, 572

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

Please See Extra Sheet.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	US,A, 3,484,162 (CLARK) 16 DECEMBER 1969 See col. 2, lines 3-15, and col. 4, line 73-col. 5, line 7.	1-3, 7-10, 18-24 1-24
Y	US,A, 3,047,507 (WINSLOW) 31 JULY 1962 See col. 2, lines 55-65.	1-24
X	JP,A, 54-158926 (RICOH KK) 19 DECEMBER 1979 See abstract	1-24

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:

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"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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Date of the actual completion of the international search	Date of mailing of the international search report
26 APRIL 1993	18 MAY 1993
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. NOT APPLICABLE	Authorized officer CHRISTINE SKANE Telephone No. (703) 308-2526

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International application No.
PCT/US93/01567

B. FIELDS SEARCHED
Electronic data bases consulted (Name of data base and where practicable terms used):

Databases searched: Chemcial Abstracts, dialog
Search terms
Eletrovis?, Electrorheo?
Dye?, Pigment?, Color?
Particle?, Particulate?