UNITED STATES PATENT OFFICE.

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CELLULOSE-NITRATE COMPOSITION.

1,309,981. Specification of Letters Patent. Patented July 15, 1919.

No Drawing. Application filed April 26, 1919. Serial No. 293,006.

To all whom it may concern:

Be it known that I, HANS T. CLARKE, a subject of the King of Great Britain, residing at Rochester, in the county of Monroe and State of New York, have invented certain new and useful Improvements in Cellulose-Nitrate Composition, of which the following is a full, clear, and exact specification.

This invention relates to a new composition of matter and a method of making the same, in which cellulose nitrate is combined or mixed with other substances, so that the resulting product can be advantageously used in the plastic and analogous arts, such, for instance, as sheet or film manufacture and varnish manufacture.

One object is to produce a composition which may be made into permanently transparent, strong and flexible sheets or film of desired thinness that are substantially waterproof, are unaffected by ordinary photographic fluids and possess the desired properties of a support for sensitive photographic coatings. Another object of my invention is to produce a composition of matter capable of easy manipulation in the plastic and film making or varnish making arts; which will not injure or be injured by the substances with which it is associated during manufacture, storage, or use. Still another object of my invention is to provide a process for compounding such composition of matter. Further objects will hereinafter appear.

I have discovered that a composition of matter having the desirable qualities hereinabove enumerated can be obtained by mixing or compounding cellulose nitrate with simple alkyl oxalates which are only slightly volatile or non-volatile at ordinary temperatures. By simple alkyl oxalates I mean substances of the type indicated by the following formulæ in which A and B represent alkyl groups containing 4 to 5 carbon atoms:

$$\begin{array}{ll} CO{-}OA & CO{-}OA \\ | & | \\ CO{-}OA & CO{-}OB \end{array}$$

There may be noted particularly dibutyl oxalate and diamyl oxalate. These may, of course, be mixed as well as used singly, and the degree of purity will depend upon the purposes to which the plastic composition is to be put. These oxalates have high boiling points and are very stable. The compounding is best performed by using a solvent common to both the cellulose nitrate and the oxalate.

As a typical useful example of the dialkyl oxalates which may be employed in carrying out my invention, the normal dibutyl oxalate is selected. It is a colorless liquid boiling under atmospheric pressure at about 243° C., and at 113° C. when under 10 mm. pressure. It has a very faint characteristic odor, and is practically insoluble in water.

In carrying out one illustration of my invention, I incorporate in 65 parts of acetone or methyl alcohol, or any mixture of the two in any proportion, 10 parts of cellulose nitrate and 2 parts of butyl oxalate. The ingredients are mixed to form a homogeneous solution or flowable mass and are filtered if desired. High boiling auxiliary softeners may be added if desired in small amounts, preferably four parts of any of the higher aliphatic alcohols or their esters, such as fusel oil, amyl acetate, butyl acetate, ethyl propionate, ethyl butyrate, etc. There may be further added, if desired, either in addition to the above mentioned high boilers or without them, such softeners as urea, castor oil, camphor, triphenyl phosphate, dibutyl sulfone, and monochlornaphthalene, 2 parts of any of these ingredients, or mixture of them, being used. The proportions of the various ingredients mentioned may be varied within certain limits. For instance, the acetone or methyl alcohol or a mixture of the two may be present in from 40 to 100 parts. The butyl oxalate may be present in from 1 to 10 parts, the higher aliphatic alcohols in from 1 to 10 parts, and the solvents last mentioned in from 1 to 10 parts.

The solution, either with or without the additional high boiling softener, is sufficiently thick and viscous to be properly flowed during sheet or film manufacture, the acetone and methyl alcohol volatilizing, but not too rapidly to impair the product. The resulting film containing the cellulose nitrate and oxalate, together with the additional high boiling softener, if the latter is used, is very flexible, transparent, and uniform, so that it can be used for any usual or preferred purpose. It is practically waterproof and unaffected by ordinary photographic chemicals.

While I have hereinabove disclosed cer-

tain compositions and processes by way of example, my invention is not limited thereto nor to the proportions given therein, as the proportions may be varied from those given and equivalent substances may be substituted without departing from the principle of my invention as defined in the appended claims.

Having thus described my invention, what I claim as new and desire to secure by Letters Patent is:

1. A composition of matter comprising cellulose nitrate and a dialkyl ester of oxalic acid in which each of the alkyl groups contains from 4 to 5 carbon atoms.

2. A composition of matter comprising cellulose nitrate and dibutyl oxalate.

3. A composition of matter comprising cellulose nitrate, a dialkyl ester of oxalic acid in which each of the alkyl groups contains from 4 to 5 carbon atoms, and a solvent common to both.

4. A composition of matter comprising cellulose nitrate, dibutyl oxalate and a solvent common to both.

5. A composition of matter comprising cellulose nitrate, a dialkyl ester of oxalic acid in which each of the alkyl groups contains from 4 to 5 carbon atoms and a solvent containing acetone and methyl alcohol.

6. A composition of matter comprising cellulose nitrate, dibutyl oxalate and a solvent containing acetone and methyl alcohol.

7. A composition of matter comprising cellulose nitrate, a dialkyl ester of oxalic acid in which each of the alkyl groups contains from 4 to 5 carbon atoms and a solvent common to both, and an organic body of only slight volatility which enhances the plasticity and flexibility of the composition.

8. A composition of matter comprising cellulose nitrate 10 parts, a mixture of acetone and methyl alcohol 40 to 100 parts, dibutyl oxalate 1 to 10 parts.

9. As an article of manufacture, a sheet of deposited or flowed cellulose nitrate containing a dialkyl ester of oxalic acid in which each of the alkyl groups contains from 4 to 5 carbon atoms.

10. As an article of manufacture, a sheet of deposited or flowed cellulose nitrate containing dibutyl oxalate.

11. As an article of manufacture, a sheet of deposited or flowed cellulose nitrate containing a dialkyl ester of oxalic acid in which each of the alkyl groups contains from 4 to 5 carbon atoms, and an organic body of slight volatility which enhances the flexibility of the sheet.

Signed at Rochester, New York, this 16th day of April, 1919.

HANS T. CLARKE.