To all whom it may concern:

Be it known that we, (1) Gustavus J. Esselein, Jr., and (2) Harry S. Mork, citizens of the United States, residing at (1) Swampscott, (2) South Boston, in the counties of (1) Essex, (2) Suffolk, and State of Massachusetts, have invented certain new and useful Improvements in Processes of Preparing Cellulose Butyrate, of which the following is a specification.

In our copending application filed April 10, 1920, Ser. No. 372,969, we have described a novel variety of cellulose butyrate and a process of preparing the same.

In a general way such process comprises a preliminary impregnation of the cellulose material with a catalyst, such as sulfuric acid, and an organic acid carrier, followed by an acylating treatment, e.g., with butyric anhydrid and butyric acid. In the specific embodiment of the process as claimed in our aforesaid application, butyric acid is used as the carrier for the catalyst employed in the impregnating step.

The present application is concerned with the use of another carrier for the catalyst, which is described but not specifically claimed in our copending application, namely acetic acid.

As stated in our copending application, we prefer in carrying out the preliminary treatment to use a bath of from 12 to 15 times the weight of the cellulose. The latter may be in any convenient form, such as cotton roving, yarn, linens, rags, etc., or in the form of wood pulp, paper, etc. It may contain a certain percentage of moisture, say 3 to 8 per cent. The proportions of the ingredients used in the preliminary bath may vary within certain limits, and the following example is to be construed purely as illustrative and not as a restriction of the scope of the invention. A suitable preliminary bath may comprise a mixture of sulfuric acid (sp. gr. 1.84) 1.0% to 5.0%, water 5.0% to 8.0%, and the balance, 94.0% to 87.0%, acetic acid.

In certain operations we have conducted, we have obtained efficient results with 4.0% of the catalyst. In general, however, the speed of the reaction depends on the amount of catalyst used. Much higher proportions of the sulphuric acid may be used, the particular amount chosen depending to a large degree on the proportions and kind of the other constituents in the preliminary bath and the acylating bath.

Similarly the water content is quite variable. Preferably from 5.0% to 9.0% is present, but the proportion may consider-ably exceed this range without harm.

After thorough impregnation of the cellulose with the catalyst, the excess of the solution containing the catalyst is removed in any suitable manner as, for example by pressing. The impregnated cellulose is then subjected to the action of the acylating bath composed of butyric anhydrid mixed with butyric acid. As an acylating bath, we have found the following very suitable, but it is to be understood that we do not limit ourselves to the specific proportions named. For each 100 parts by weight of cellulose (calculated on the untreated cellulose), we may employ 405 parts butyric anhydrid (90.0%) and 400 parts butyric acid.

If the proportions of the various materials have been properly regulated, there will be a rise in temperature which can be readily controlled and which need not exceed 40° or 50° C. The reaction is allowed to proceed until the product has the desired solubility. If the sulfuric acid employed in the preliminary bath has been used in relatively small proportion, it is advisable, after a test sample shows good solubility in chloroform, to add sulfuric acid to the butyrating mixture. For this purpose a mixture made up substantially of 5.0% sulfuric acid (sp. gr. 1.84), calculated on the weight of the cellulose employed, and from 3.0% to 5.0% of water by volume of the butyrating bath is suitable; but the proportion of acid added may of course be varied. This accelerates the transformation and produces the desired solubility in benzol and denatured alcohol with less degradation of the product than if the reaction is allowed to proceed for a longer time without this addition.

As stated in the original application the product is soluble in chloroform, acetylene tetrachloride, acetone, ethyl acetate, 90% carbolic acid, hot denatured alcohol, hot benzol, and at room temperature in mixtures of alcohol, either ethyl or methyl, with benzol, containing from 50.0% to 95.0% by volume of benzol. It is also soluble in mixtures of 70.0% carbon tetrachloride and 25.0% by
volume of denatured alcohol, and in hot solvent naphtha. It is not soluble in normal butyl alcohol, solvent naphtha or carbon tetrachloride at room temperature.

5 We claim:—

1. The process of preparing cellulose butyrate which comprises subjecting cellulose to a preliminary impregnating bath containing an acid catalyst, a carrier therefor comprising acetic acid, and water, and butyrating the impregnated cellulose.

2. The process of preparing cellulose butyrate which comprises impregnating cellulose in a bath containing substantially 1.0% to 5.0% sulfuric acid, the remainder of the bath consisting of acetic acid and water, and butyrating the impregnated cellulose.

3. The process of preparing cellulose butyrate which comprises impregnating cellulose in a bath containing substantially 1.0% to 5.0% sulfuric acid, 5.0% to 8.0% water and the remainder acetic acid, and butyrating the impregnated cellulose.

4. The process of preparing cellulose butyrate which comprises incorporating with cellulose an acid catalyst associated with acetic acid as a carrier, and subjecting the treated cellulose to an acylating bath containing butyric anhydrid and butyric acid.

5. The process of preparing cellulose butyrate which comprises subjecting cellulose to a preliminary impregnating bath comprising a catalyst and a carrier therefor comprising acetic acid, and water, and subjecting the impregnated cellulose to an acylating bath containing substantially 400 parts butyric acid and 465 parts butyric anhydrid (90%) for each 100 parts by weight of cellulose.

6. The process of preparing cellulose butyrate which comprises impregnating cellulose in a bath containing substantially 1.0% to 5.0% sulfuric acid, 5.0% to 8.0% water and the remainder acetic acid, and subjecting the impregnated cellulose to an acylating bath containing substantially 465 parts of butyric anhydrid (90%) and 400 parts butyric acid for each 100 parts by weight of cellulose.

7. As a new product, a cellulose butyrate such as can be produced by the described process comprising impregnating cellulose in a bath containing substantially 1.0% to 5.0% sulfuric acid, 5.0% to 8.0% water and the remainder acetic acid, and subjecting the impregnated cellulose to an acylating bath containing substantially 465 parts of butyric anhydrid (90%) and 400 parts butyric acid for each 100 parts by weight of cellulose.

In testimony whereof, we affix our signatures.

GUSTAVUS J. ESSELEN, Jr.
HARRY S. MORK.