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ADHESIVES

Paul B. Davidson, Wilkinsburg, Pa., and Julius R. Adams, Westfield, Mass., assignors to Old Colony Envelope Company, Westfield, Mass., a corporation of Massachusetts

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This invention relates to improvements in adhesives and is more particularly related to so-called gums for use in connection with envelopes or other devices where it is desired to adhesively secure together adjacently-disposed surfaces, especially paper surfaces as in the case of envelopes.

This application is a continuation in part of an application Ser. No. 202,882 filed by us on April 19, 1938, issued as Patent No. 2,241,700.

It is a principal object of the invention to provide an adhesive for use with paper which is so constructed chemically that it will not discolor or otherwise deteriorate with age. That is to say, we have conceived an adhesive which will not have the tendency to discolor regardless of climate or other conditions under which the envelopes or other articles are stored.

It has always heretofore been a serious difficulty in the envelope industry that the envelopes, after storage on shelves or the like, will become discolored due to reaction between the gum or adhesive and elements in the paper. It appears that the cause of discoloration is the formation of colored or humus-like substances due to reaction between degraded protein which is necessarily present in any ordinarily sized paper and the reducing sugars which are present in any ordinary paper adhesive.

A further object of the invention is to provide an envelope adhesive which has special properties and characteristics particularly adapting them for use in the manufacture of envelopes. It is the usual practice in the envelope industry to apply the gum thereto by means of what is called an envelope machine and it will be appreciated that in order that application of the gum be carried on efficiently and economically the adhesive must remain sufficiently fluid and non-pasty for an ample length of time. The gums of this invention not only have the non-discoloration properties above-mentioned but also they are properly fluid for use in the envelope machine.

After considerable research we have come to the conclusion that the problem of discoloration can be solved. Heretofore, to our knowledge, no one has ever even attempted to deal with the matter of discoloration, much less provide an adhesive adapted to eliminate it. Nor is there any prior art which suggests any previous treatment or procedure to provide an envelope gum which is both sufficiently fluid and not capable of causing discoloration.

Our invention consists broadly in providing paper adhesives which have as a base a certain amount of partially-dextrinized starch and contain one or more of a group of fluidifying substances which are capable of both sufficiently inhibiting objectionable gel formation and preventing discoloration. That is to say, the composition is such that it is properly fluid to permit efficient application to the paper by the envelope machines and it is also such that it will not, after application to the paper, tend to react with more or less degraded protein in the paper sizings so as to form colored compounds which are objectionable.

After considerable experiment we have found that any gums or adhesives which contain appreciably more than 2% by weight of reducing sugars do react unfavorably with the protein in ordinary paper sizings so as to produce discoloration. In other words, we provide an adhesive composition which is so constructed chemically as to have a total reducing-sugar content which is preferably not more than 2% by weight of the gum and we accomplish this by a combination of partially-dextrinized starch and a fluidifying agent which is calculated to produce the desired effect.

Careful and scientifically conducted experiments have clearly shown us that when the reducing-sugar content of the gum does rise appreciably above the percentage mentioned, there is, inevitably, discoloration. For example, a number of pieces of paper, all the same kind, were stuck together with various adhesives which have been heretofore known and with adhesives of this invention which are about to be described.

These pieces were aged by an accelerated ageing test which had been standardized against natural conditions. This test consisted of placing the pieces in test tubes, sealing them, and placing the tubes in an oven regulated to 60° C. for 48 hours. Discoloration appeared in the pieces which were not gummed with the adhesives of this invention so we proceeded to determine the reducing sugars in all of the sample gums.

This was done by diluting a sample of each gum with 100 cc. of water and precipitating the dextrins by adding 100 cc. of 98% ethanol, according to the method described as the "Starch Tentative" on page 385 of the third edition of "Official and Tentative Methods of Analysis of the Association of Official Agricultural Chemists."

The dextrins were allowed to settle out of dispersion for 48 hours and then samples of the

clear supernatent liquid were taken for the reducing sugar determination in accordance with the common and well-known method described as the "Defren's Method" on page 74 of the second edition of "Methods of Organic Analysis" by Dr. H. C. Sherman.

However, for the gravimetric method for determining the reduced copper in the "Defren's Method," we substituted the more accurate volumetric thiosulfate method which is the official method described on page 380 of the text of the Association of Official Agricultural Chemists mentioned above. It was clear from the results that the particular adhesives embodying the features of this invention did not show discoloration because the reducing sugar content was below 2%, all other things having been kept equal.

We have further found that there are particular types of dextrin or starch which are most suitable for accomplishing our purpose. While, as a general rule, best results can be obtained if the reducing sugar content of the dextrin itself before cooking is below 2%, it is necessary that the final cooked gum contain less than 2% of reducing sugar to completely avoid discoloration.

That is to say, the hydrolysis that takes place during the normal cooking of the gum may objectionably increase the reducing sugar content even though the content of the original dextrin was low. Accordingly, we preferably use a partially-dextrinized starch which is of such a composition that when it is cooked for about thirty minutes at a temperature of around 98° C. with an approximately 40% concentrated solution of acetic acid, it will have, upon cooling, a reducing sugar content of less than, or at least not appreciably more than, 2% by weight.

Another example of a preferred dextrin is one which, when cooked for about thirty minutes at a temperature of about 85° C. with the same acetic acid, will not have a reducing sugar content of more than 2% by weight. That is to say, of all the variously dextrinized starches which are commercially available those which have the just-referred-to qualifications are most suitable for the practice of this invention.

When using the terms "dextrin" or "partially-dextrinized starch" hereafter, we mean just such substances as above defined. We found that while such mixtures as just described might have the desired low reducing sugar content, they were not sufficiently fluid for application by the machines so that it was incumbent upon us to fluidify the mixtures in a way which would not increase appreciably the sugar content. As stated, the common way to fluidify heretofore has been to add more sugar; this, of course, increases the sugar content so that the gum would have all of the above-mentioned objections.

Such substances as we have found to produce the desired result include the amides of monocarboxylic acid containing from 2 to 5 carbon atoms inclusive. We desire to here point out that we are concerned with amido compounds, such as acetamide, for example, which are to be distinguished from the so-called "amido" acids heretofore disclosed in a United States Letters Patent #974,448 of 1910 to one Supf. According to Rule 5 adopted by the nomenclature committee of the joint congress of the British Chemical Society and the American Chemical Society held in 1923: "The names of the groups NH_2, NHR, NR_2, NH or NR should end in '-ido' only when they are substituents in an acid group, otherwise in '-ino'."

Other agents which we have found suitable are propianamide and butyramide.

That is, we are here concerned with amides or salts of acids, as contrasted with what are now called "amino" acids and which in Supf's time were referred to as "amido acids." To put it another way, while in 1910, for example, the usual definition for an amido group was one where an NH_2 group replaces a hydrogen of the hydrocarbon, today such terminology indicates the replacing the OH of a carboxylic acid to form an amido compound. We are concerned with present-day amido compounds rather than what was synonymous in 1910 with an amino group.

According to the preferred form of the method of this invention, water and the partially dextrinized starch are mixed intimately. If desired, a volatile acid, such as acetic, may be introduced to aid in the hydrolysis of the starch and the mixture is then agitated so that the ingredients thoroughly blend.

The composition is then heated in any well-known manner to within a range of preferably from 160° to 212° F., which temperature is preferably maintained for from 20 to 60 minutes. The length of time during which the gum is cooked depending, of course, upon the size of the batch, the temperature, the equipment used, and various other factors.

It is desirable that the mixture be cooked only until hydrolysis to the desired degree of dextrinization is complete and not for such a length of time that the reducing sugar content be substantially increased. The fluidifying agent may be now added but we do not wish to be limited to any specific order in which the above ingredients may be added.

As final steps in the process the gum is preferably chilled to about 100° F., or below, after which the mixture is ready to be drawn.

While the proportions of the above ingredients may be greatly varied, very favorable results have been obtained when staying within the following approximate ranges of proportions by weight:

	Per cent
Water	10 to 18
Partially dextrinized starch	50 to 62
Fluidifying agent	3 to 10

If acetic acid or some other volatile acid is added, about 10 to 20% thereof may be used. If it is desired to include glycerine, about 4 to 10% thereof may be used.

While we have described the invention in great detail and with respect to the present preferred form thereof, it is not desired to be limited thereto since many changes and modifications may be made therein without departing from the spirit and scope of the invention. What it is desired to claim and secure by Letters Patent of the United States is:

1. As a new article of manufacture, an envelope adhesive having a reducing sugar content of not more than substantially 2% of the weight thereof, comprising in combination, partially-dextrinized starch and an amido compound adapted to fluidify the adhesive for a sufficient length of time to permit its proper application to the envelopes by the envelope machines and to render the adhesive incapable of causing discoloration after being applied to the envelopes, said amido compound consisting of acetamide.

2. As a new article of manufacture, an envelope adhesive having a reducing sugar content of not more than substantially 2% of the weight thereof, comprising in combination, partially-dextrinized starch and an amido compound adapted to fluidify the adhesive for a sufficient length of time to permit its proper application to the envelopes by the envelope machines and to render the adhesive incapable of causing discoloration after being applied to the envelopes, said compound consisting of an amide having between 2 and 5 carbon atoms inclusive.

3. As a new article of manufacture, a paper-envelope adhesive of the character described consisting of, a certain amount of partially-dextrinized starch, water, and an amide of monocarboxylic acid containing from two to five carbon atoms inclusive, said starch, water, and amide being in such relative proportions that the total reducing sugar content of the adhesive is limited to about 2% of the weight thereof.

4. As a new article of manufacture, a paper-envelope adhesive of the character described consisting of, a certain amount of partially-dextrinized starch, water, and an amido compound of the group consisting of acetamide, propianamide and butyramide, said starch, water and compound being in such relative proportions that the total reducing sugar content of the adhesive is limited to about 2% of the weight thereof.

PAUL B. DAVIDSON.
JULIUS R. ADAMS.