PEANUT PROTEIN HYDRATES 1/

Utilization as Tacky and Remoistening Adhesives

THE peptization characteristics of the nitrogenous constituents of peanut and soybean meals are similar (*5*), and solutions of proteins isolated from these meals have similar viscosity relations (*3*). These and related investigations indicate that peanut meal and isolated peanut proteins have the same industrial applications as soybean meal and proteins. These applications have been limited largely to those to which casein is best adapted—namely, the production of plywood glues, paper coatings and sizes, cold water paints, and similar products where tacky or remoistening types of adhesives are not required or would be disadvantageous.

There is, however, a large field in which the applied wet glue film must be tacky in order to hold two surfaces together until the glue film dries to form a more permanent bond. For example, paper-box makers and bookbinders use large quantities of tacky adhesives, called "flexible glues", which are ordinarily mixtures of bone or hide glue with sufficient glycerol, sorbitol, or sugars to give flexibility to the dry glue film.

There is also a large adhesive field in which a dried glue coating must develop adhesive properties and form a bond when it is moistened with water. Such varieties are called "remoistening" or "gumming" adhesives. Large amounts of gummed paper suitable for labels, stamps, and envelope seals are prepared with dextrin adhesives. Animal glues are employed for making gummed tape or fabric for sealing the heavier types of cartons. For this use the remoistened adhesive coating must develop strong adhesive properties and set up rapidly to form a bond. About 20 million pounds of bone glue and an estimated 25 million pounds of dextrins are used annually for gumming purposes.

These adhesives must have tacky and/or remoistening properties, and they must possess a pH value which is neither strongly acid nor alkaline. This paper describes the preparation from peanut proteins of adhesives which meet these requirements. A description of peanut and soybean hydrates from which the tacky and remoistening adhesives are prepared was reported in a previous article (*1*).

R. S. BURNETT, E. D. PARKER, AND E. J. ROBERTS
Southern Regional Research Laboratory, U. S. Department of Agriculture, New Orleans, La.

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DEWATERED PEANUT PROTEIN CURDS

Protein curds obtained by precipitation at pH 5.0 contain practically all the protein which was in the peanut meal extract, since this pH value lies within the range of minimum solubility of the peanut proteins (*2*, *5*). Furthermore, the curds obtained by precipitations at pH 5.0 can be dewatered more readily to yield the protein hydrate than can curds obtained by precipitation at pH 4.5. Dewatering is accomplished by heating the curds to about 50° C., at which temperature the jelly-like particles of curd coalesce and exclude water present in excess of that bound by the protein molecules. This was discussed in a previous publication (*1*).

There are two advantages of dewatering the protein curds before drying. First, by lowering the water content to about 42.5 %, this method reduces the cost of drying the filtered curds which originally may contain as much as 80% water. Second, it results in dried protein of uniform ash content, since the water content of two batches of dewatered curds prepared by the same method is approximately the same. Any variation in the amount of water retained by the curds would produce a variation in the soluble nonprotein content of the dried protein.

Glues have been prepared from peanut protein, isolated from both solvent-extracted and hydraulic-pressed peanut meals, which are suitable for making gummed tape and paper and for making flexible glues of the type used by the setup paper-box maker and the bookbinder. Preliminary investigations have shown that gummed tapes with relatively good adhesive strengths can be prepared from peanut protein glues. Peanut protein flexible glues are relatively free of difficulties encountered with certain other types because they can be applied cold and become tacky immediately. Readily soluble glues can be prepared from isoelectric peanut protein curds by neutralizing the curds before they are dried. Dewatering peanut protein curds to the amount of water in the hydrate reduces drying costs and provides a glue of relatively uniform ash content. The characteristics of these peanut protein glues described, particularly rewettability, tackiness, and flow properties, render them suitable for certain gluing purposes for which vegetable proteins have previously been considered unsuitable.

PREPARATION OF PROTEINS FOR ADHESIVES

Dried peanut proteins obtained from curds precipitated at pH 4.5 to 5.0 must be dissolved at pH values between 6.0 and 9.0 in order to obtain sol hydrates which have viscosities suitable for use as adhesives of the type under discussion. This procedure is difficult and time consuming, and requires the use of control instruments and supervision by technical personnel, which are generally unavailable even in factories where adhesives of this type are used on a large scale. The difficulties may be circumvented by neutralizing the dewatered, acidic curds with sodium hydroxide solution before drying. The dried protein is readily soluble, provided water is not present in excess of the amount which can be bound by the protein at any given pH value. These "preneutralized" proteins are referred to here as glues. They have aptly been called "goober glues" (*4*).

Two series of experimental glues were prepared for gumming purposes. One was made from protein curds isolated from solvent-extracted peanut meal; the other was made from protein curds isolated from a hydraulic-pressed meal prepared in the pilot plant under mild cooking conditions—that is, at meal temperatures not exceeding 215° to 218° F. Details of the preparation of the glues are given in Table I.

In each case the peanut protein was extracted from the meal at pH 7.7 with aqueous sodium hydroxide, the meal to solvent ratio being 1 to 10. Since it is not necessary to clarify the protein extract completely prior to precipitation of the protein, if the latter is to be used for preparation of adhesives, the extract and insoluble residue were separated by a solid basket centrifuge. The extract was then allowed to stand about an hour to permit the small amount of suspended matter to settle, the supernatant solution was separated, and the protein was precipitated by the addition of sufficient sulfur dioxide to bring the pH to 5.1.

The curds obtained from the two meals were dewatered, and each dewatered curd was divided into three portions. Each of the three portions was adjusted to a different pH (Table I), and the curds were finally dried in an oven at 50° C. These neutralized curds are difficult to dry, since they consist of thick solutions which cannot be subjected to temperatures above 50° to 60° C. without affecting the protein, as indicated by a change in the viscosity characteristics of solutions prepared from the protein. It is probable that this material can be efficiently spray-dried or vacuum-drum-dried. However, the neutralized, dewatered peanut protein curds can be used advantageously without prior drying when the glue is used by the manufacturer or when the cost of shipping the 30–35% excess water in the glue is equal to, or less than, the cost of drying. Plasticizer, when needed, may be added by either the manufacturer or by the user. Most of the animal flexible glues now on the market contain 25 to 50% water and need only to be diluted and melted before being used. A flexible glue made from neutralized and plasticized peanut protein curds would need only to be diluted before being used.

GUMMED TAPE

The six peanut glues described above were used to make gummed tape in a gummed paper factory. Regular commercial equipment was used for all steps in the process, with the exception of the glue mixer, which was too large for the experimental material available. The glue solutions were prepared by soaking 6 parts of glue with 4 parts of water for 1 hour. The thick pastes thus obtained were then diluted with water until they appeared to be of suitable consistency for application. After dilution, these solutions contained from 40 to 45% protein. No viscometer was available at the plant to measure the resulting solutions, but from previous work it was known that peanut protein solutions having this range of concentration usually have viscosities of approximately 30 to 40 poises.

Each glue solution was poured on a traveling belt of kraft gumming paper just before the paper passed under a doctor blade which had been set to give approximately the desired coating thickness. The gummed paper was passed through drying tunnels where the glue film was dried by a blast of warm air. The dried gummed paper was made into a single roll, which was finally cut into rolls of smaller widths, including 2-inch rolls for adhesion tests and 3-inch rolls for gluing cartons.

The device and method for making the adhesion tests are described as follows: The instrument was a manually operated McLaurin gummed tape tester which "consists of two tables about 6 × 8 inches placed side by side. Each table is hinged at the outer edge on the underside so that the adjacent ends are free to move in an arc. In testing, these are moved by the action

Table I. Preparation and Tests of Peanut Protein Glues[a] and Gummed Tape

Glue No.	Meal Used as Source of Protein	pH of Extn.	pH of Pptn.	pH of Dewatered Curd before Drying	pH of Dried Glue	Coating Wt.[b]	Average McLaurin Adhesive Strength	Practical Test by Taping No. 102 Cartons
1	Solvent-extd.	7.7	5.1	7.3	7.1	9.1	75	Better than 2 or 3
2				8.6	8.2	10.0	36	Trifle slow setting
3				9.6	8.8	12.2	11	Trifle slow setting
4	Hydraulic-pressed	7.7	5.1	7.3	7.3	11.6	66	Little slower setting than 5 & 6
5				8.4	8.0	10.7	72	Better tack than any of others
6				9.5	9.3	10.0	60	Same as 5 except slight tendency of tape to let go at edges

[a] For comparison, gummed tape prepared with animal glue had a coating weight of 7.0 and an adhesive strength of 92.
[b] "Folio base", pounds per ream of 500 sheets, 17 × 22 inches.

of a cam on the end of a weighted level arm. The action of the tables is intended to duplicate the action of the flaps of a corrugated case after they have been closed and then the pressure removed, allowing them to spring back to the partially open position. In addition to the above, the machine is equipped with suitable trip levers, graduated scale, brackets to hold the test specimen, and frame to hold the entire assembly. In making the tests a sheet of the test paper about 6 × 10 inches is placed across the top of the two tables, and the holding clamps are tightened down securing the ends of the sheet. The test sheet is then slit through the center or directly over the position where the two tables come together. A piece of gummed tape to be tested is moistened and then placed on this test sheet, and after the proper interval of time the machine is tripped and the adhesive strength taken directly from the scale reading" (*6*).

Further details are given in the paper from which the preceding is quoted and in the recently issued TAPPI Tentative Standard for testing gummed paper tape (*7*).

Results of the tests conducted on the tapes gummed with peanut protein glues are given in Table I. Results of a practical test made by actually gluing cartons with the experimental gummed tapes are also given in this table. In general, results of the carton gluing tests agree with those of the McLaurin tests.

Results obtained with peanut protein glue approach closely those obtained with animal glue, but it would be unsafe to conclude that any one of the peanut glues was superior to another, as a large number of factors in this preliminary test were not controlled, and any one of them may have adversely affected the results. For example, differences in the viscosities of the glue solutions may have caused variations in the amount of glue absorbed by the paper, which would, in turn, cause variations in the amount of available glue on the surface of the tape. The results may have been affected also by variations in the glue coating applied; too much glue may be as detrimental as too little.

Although all the advantages and disadvantages of peanut protein glues as compared with other gumming adhesives have not yet been investigated, the following advantages of peanut protein glues are apparent: uniformity of product; superiority of color, especially when produced from special varieties of peanuts (*2*), hence preferable for gumming white paper; less hygroscopicity than is found in animal glues or dextrins and, consequently, less tendency of the gummed paper to stick in the roll or stack when it is stored in hot humid atmospheres; highly sanitary product, which is especially important where the glue is remoistened with saliva.

A disadvantage of the peanut protein glues tested so far is the tendency for the tape prepared with these glues to blister after it has been on the carton for some time. This tendency is also exhibited by tapes gummed with dextrins and is disadvantageous primarily from the point of view of appearance rather than of utility.

It is evident, then, that a peanut protein gummed tape with good adhesive strength can be made from the proteins of both solvent-extracted and hydraulic-pressed peanut meals. A systematic investigation of all the factors involved will probably result in an increase in the adhesive strength of the tapes until they are comparable with those made with animal glue.

Peanut glues in their present state of development, however, are suitable for gumming purposes where dextrins are now employed and for many purposes where animal glue is used.

FLEXIBLE OR NONWARP GLUES

Solutions of the glues used in the gummed tape tests were employed in experimental gluing of the paper wraps on setup paper boxes. Candy boxes were used for the tests, and the glue solutions were applied to the paper wraps by means of a small table gummer, ordinarily employed by the bookbinder and paper-box maker in hand operations. These boxes showed a tendency to warp when stored in a relatively dry atmosphere (32% relative humidity). However, when 50% sorbitol (dry glue basis) was added to the glue solution, the boxes did not warp when stored under the same conditions. The warping tendency of the boxes prepared with unplasticized peanut glues was much less than that of boxes prepared with unplasticized animal glue. Accordingly, peanut nonwarp glues may be expected to require less plasticizer than animal glues.

An important advantage of flexible peanut protein glues, as compared with flexible animal glues, lies in the fact that peanut protein glues can be applied cold, are tacky as soon as they are applied, and remain tacky long enough to complete a given gluing job. Animal flexible glues must be applied hot and are not tacky until they have cooled sufficiently to start gelling. Furthermore, animal glues allowed to cool too much lose their tackiness, owing to complete gelling of the glue film. For this reason users of flexible animal glues experience considerable difficulty whenever changes occur in the atmospheric temperature or relative humidity of sufficient magnitude to affect the rate of cooling of the glue film.

Although no extensive investigation has been made with soybean protein, protein hydrates having properties and uses similar to those of peanut protein can probably be produced from solvent-extracted soybean meal.

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LITERATURE CITED

(1) Burnett, R. S., Ind. Eng. Chem., 37, 861 (1945).
(2) Burnett, R. S., and Fontaine, T. D., *Ibid.*, 36, 284–8 (1944).
(3) Burnett, R. S., Roberts, E. J., and Parker, E. D., *Ibid.*, 37, 276–81 (1945).
(4) Chapman, P. W., *Atlanta Journal*, Mag. Sect., p. 14, June 25, 1944.
(5) Fontaine, T. D., and Burnett, R. S., Ind. Eng. Chem., 36, 164–7 (1944).
(6) Harnden, G. H., A.S.T.M. *Bull.*, 98, 23–7 (1939).
(7) Tech. Assoc. Pulp Paper Ind., Tentative Standard T463 m–43, *Paper Trade J.*, TAPPI Section 116, 267 (1943).