PULP- AND PAPER-MAKING POTENTIAL OF PEANUT HULL WASTE IN BLENDS WITH SOFTWOOD PULP[1]

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ABSTRACT

The use of peanut hulls as a raw material source for the manufacture of cellulose fiber products was evaluated by abbreviated kraft and soda pulping processes. Several types of experimental paper products were made from the unbleached and partially bleached pulps. Breaking lengths and tear factors were determined for handsheets made with peanut hull pulp alone and with mixtures of peanut hull and softwood pulp. Some handsheets blended from peanut hull and wood fibers were stronger than those made from pure wood pulp. In view of the results obtained, it appears feasible to employ peanut hull waste to extend the fiber raw material basis of pulping processes without sacrificing paper quality.

Additional keywords: Agricultural residues, kraft, nonwood fibers, paper tests, soda.

INTRODUCTION

The increasing demand for paper products has initiated a widespread search for fibrous raw materials other than wood. Next to secondary fibers, annual crops and agricultural residues also offer possibilities to supplement wood as a raw material source for paper production. The reluctance on the part of industry to employ such nonwood materials stems partly from seasonal variations and limitations in supply flows, from spoilage in months-long storage, or simply from detraction from process efficiency or product performance. As technology progresses in an evolutionary fashion, the pulp industry will have to learn to use raw materials that have not been used before. Peanut hulls seem to offer one particularly well-suited escape from wood supply shortage since they become available as waste products from shelling operations at locally concentrated sites and on an almost year-round basis. The cultivation and processing of peanuts is an extensive industry in the southeastern portion of the United States. Among the by-products of this industry are the hulls removed at the processing plant. About 400,000 tons per year of these shells are obtainable in the Southeast, most of which at present are either burned or sold as mulch or as feed for animals. It appears, therefore, that there are good grounds for attempting to find an industrial use for these hulls.

In the past, peanut hulls and related agricultural waste products have been used for the manufacture of particle and insulation boards (Chittenden and Palmer 1964 and 1965; Jain et al. 1964) and pulps (Lynch and Goss 1930; Govil 1960), among others.

The cellulosic substances, which comprise aproximately 46% by weight of the peanut hull and fibers measuring 1.7 mm long on the average (Lynch and Goss 1930), could be used for the production of pulp and paper. Efforts to demonstrate this use for peanut hulls have continued for

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TABLE 1. *Composition of Peanut Hulls vs. Wood* (Pinus resinosa).

Component	Percent		Reference[1]
	Peanut Hulls	Red Pine	
Extractives			
Alcohol	3.2-3.3	9.7[2]	Lynch and Goss 1930
Water	7.3-12.7	1.8[3]	"
Ether	0.5-0.6		"
1% NaOH Solubles	21.5-21.6		"
2% H_2SO_4 Solubles	25.5-25.6		"
Ash: Total	1.6-4.6	0.2	Lynch and Goss 1930
SiO_2 in Ash	14.7-16.1		Govil 1960 Govil 1960
Nitrogen	0.9-1.8		Lynch and Goss 1930
Protein	5.6-7.9		"
Lignin	33.4-33.7	23.4	"
Pentosans	18.5-19.4		"
Hemicellulose		15.1	
Cellulose:			
Cross-Bevan Method	36.6 47.2-51.0		Govil 1960 Lynch and Goss 1930
Sieber-Walter Method	43.5-45.0		"
Jansen-Norman Method	35.2		Govil 1960
α-cellulose		47.8	

1) References refer to peanut hull composition; values for wood were obtained from Browning (1963)

2) Alcohol-benzene

3) After extraction with alcohol-benzene

many years. In addition to some special pulping processes that have been designed especially for use with nonwood raw materials (see Institute of Paper Chemistry 1970; Rydholm 1965), such as multistage pulping with nitric acid (Mills 1969 and 1970), conventional pulping processes based on alkaline and neutral liquors have been proposed (Lynch and Goss 1930; Govil 1960). Lynch and Goss (1930) used soda, kraft, and neutral sulfite processes on peanut hulls, but obtained only "unbleachable" or "bleachable poor pulp" in low yields. Papermaking efforts were quoted from the Forest Products Laboratory, which found "test sheets with equal parts of peanut hull pulp and wood pulp . . . to be weak." Another study by Govil (1960) describes the soda process and high temperature as unsuited for the pulping of peanut hulls; in contrast, the sulfate method was used, and yields of 30–35% were obtained. No paper was made from the pulp.

It was the objective of this investigation to study the pulp- and papermaking properties of peanut hulls using conventional soda and kraft processes that may permit the use of hulls as substitutes for and additives to wood chips. Since the fibers are rather short, it would not be expected that a paper product made solely of peanut hulls would be of the same quality as paper made of wood pulp. However, the potential value of peanut hull pulp lies in blending it with wood pulp. Furthermore, numerous pulp and paper mills are located in the southeastern states that afford ideal access to the supply of peanut hulls, and shipping costs will thus be minimized.

EXPERIMENTAL

The major components of peanut hulls have been analyzed following procedures designed for wood. In Table 1 some of the analytical data for peanut hulls that are available in the literature have been compiled and are compared to values obtained with wood. The surprisingly high

TABLE 2. *Results of Pulping and Bleaching of Peanut Hulls.*

Sulfidity (%)	NaOH (g)	Na_2S (g)	Pulp Yield (%)	Permanganate No. Unbl.	Permanganate No. Bl.	Brightness Unbl.	Brightness Bl.	Freeness Unbl.	Freeness Bl.	Amount of Shives
0	193.5	0	57.3	25.7	1.4	17.9	59.9	200	495	low - none
15	164.5	28.3	53.8	22.5	6.7	19.7	57.6	270	360	low - medium
30	135.6	56.6	55.0	24.7	2.7	14.1	57.2	280	305	medium - high
50	96.7	94.3	47.5	28.4	16.7	12.9	28.6	290	295	very high (unbleached: appr. 50% shives)

content of lignin in peanut hulls might reflect the condensation of proteins and lignin under the influence of the lignin-determining treatment (sulfuric acid), as pointed out by Lai and Sarkanen (1971). However, the lignin content is of secondary importance since pulping aims at complete fiber separation combined with good bleachability rather than at complete delignification. Cellulose and hemicellulose contents of peanut hulls seem to be very similar to those of wood tissues.

Pulping procedure

The liberation of fibers from peanut hulls was carried out with alkaline liquors containing varying quantities of sulfides. Sodium hydroxide and sodium sulfide were the active chemicals used. The degree of sulfidity, which is determined by the concentration of sodium sulfide, was varied from 0 to 50%. For each test, 750 g of oven-dry (OD) peanut hulls were pulped, and the total amount of active chemicals (referring to NaOH + Na_2S, expressed as Na_2O) was set at 20% of the weight of the hulls or 150 g. The chemicals were dissolved in 2.5 liters of water to which the hulls were then added. The contents were placed in an autoclave and cooked at 180 C for 1½ hr after ½ hr heatup time. Preliminary tests had shown that these were optimum conditions to obtain pulps that disintegrate well with permanganate numbers of about 25. Four tests were conducted in which the sulfidities were set at 0%, 15%, 30%, and 50%. The pulps were filtered through paper, washed thoroughly, and disintegrated for 10 min. The results are compiled in Table 2.

Bleaching

A partial bleaching process consisting of the following three stages was carried out on the unbleached peanut hull pulps (PHP): 1) chlorination, 2) extraction with alkali, 3) hypochlorite treatment. The length and temperature of the treatments were kept as low as possible in order to avoid excessive degradation of cellulose.

For each test, 45 g of OD unbleached pulp was used. In the chlorination stage, the amount of chlorine used was 5.2% of the weight of the pulp; pulp consistency was 3%. The mixture was left for 45 min at room temperature; afterwards, the suspension was filtered and washed until it was neutral. The amount of sodium hydroxide used in the alkaline extraction stage was 2.5% of the OD pulp; pulp consistency was 10%. The alkali was allowed to react with the pulp at 75 C for 1 hr under agitation. Afterwards, the suspension was filtered and washed free from alkali. In the final stage, the amount of active chlorine in the hypochlorite solution was 1.8% of the OD pulp at a pulp consistency of 10%. The *pH* value of the suspension was periodically checked and kept between 10.4–11.2. The suspension was heated to 35 C and kept at this temperature for 2.5 hr. Afterwards, the suspension was filtered and washed with water. Results are summarized in Table 2.

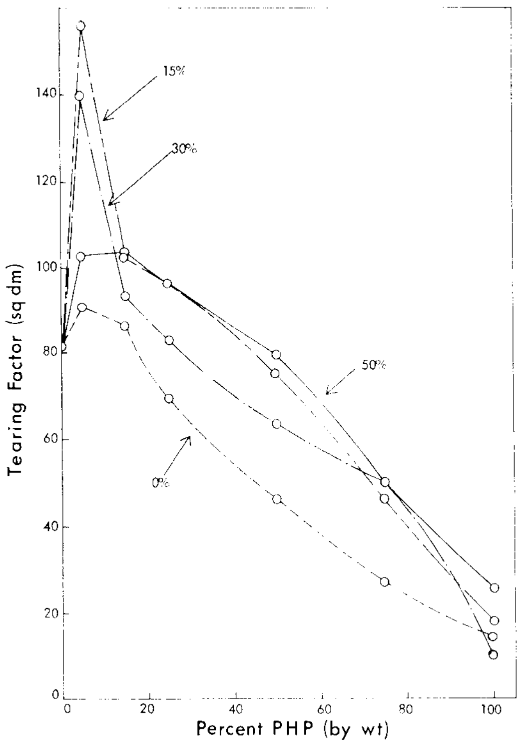

Fig. 1. Tearing Factors of Blended Handsheets Using Unbleached Peanut Hull Pulps.

Formation and testing of handsheets

Standard 1.2 g handsheets were made from unbleached and partially bleached PHP following TAPPI standard T205, and their physical properties were determined. Tear factor and breaking length data were obtained by using an Elmendorff tear tester and Instron tensile apparatus, respectively. Testing procedures used were according to TAPPI standard T220. Also, blended handsheets composed of 0%, 5%, 15%, 25%, 50%, and 75% by weight PHP and bleached softwood pulp (Special Pulp, Testing-Blotting, by: SORG PAPERS, Copco Papers, Columbus, Ohio, Distributors, manufactured in accordance with TAPPI standard T205 m-50) were made and tested. For the tearing tests, the data were obtained by testing either four or five handsheets together and, usually, by making five tears on each sample whose values were averaged. Five or six strips of sample

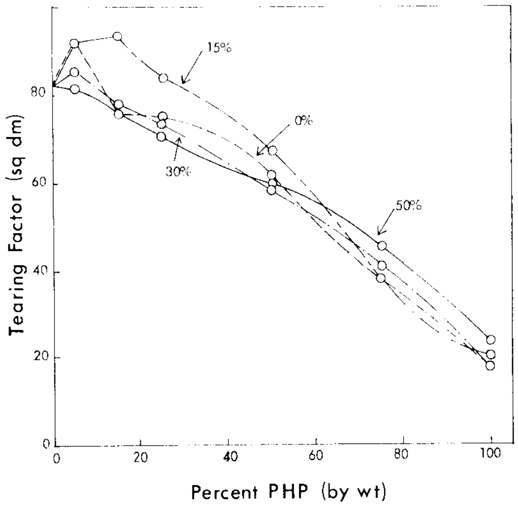

Fig. 2. Tearing Factors of Blended Handsheets Using Bleached Peanut Hull pulps.

paper were used separately in determining the tensile strengths, and the average reading was determined. The Canadian Standard Freeness test was performed on bleached and unbleached PHP in accordance with TAPPI standard T227. Brightness was determined following the procedure outlined in TAPPI standard T217.

The 50% sulfidity unbleached kraft pulp was unsuited for handsheet formation without prior screening. Therefore, the 50% sulfidity pulp was screened in order to remove undelignified particles (shives), and subsequently handsheets were made from the screened pulp only. In forming blended handsheets, the unbleached PHP obtained from the 30% and 50% sulfidity cooks were screened, while the remaining pulps were used as obtained from the laboratory tests.

RESULTS AND DISCUSSION

The pulping of peanut hulls with alkaline liquors at various sulfidity levels seems to produce pulps in far higher yields than has been reported previously (Lynch and Goss 1930; Govil 1960). As indicated in Table 2, 53–58% of the hulls were obtained as pulp if the sulfidity was varied between 0 and 30%. Relatively short cooking periods of 1½ hr, plus ½ hr heatup time were suf-

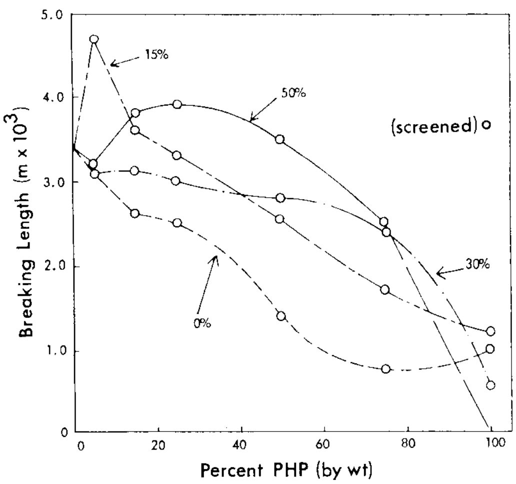

FIG. 3. Breaking Length of Blended Handsheets Using Unbleached Peanut Hull Pulps.

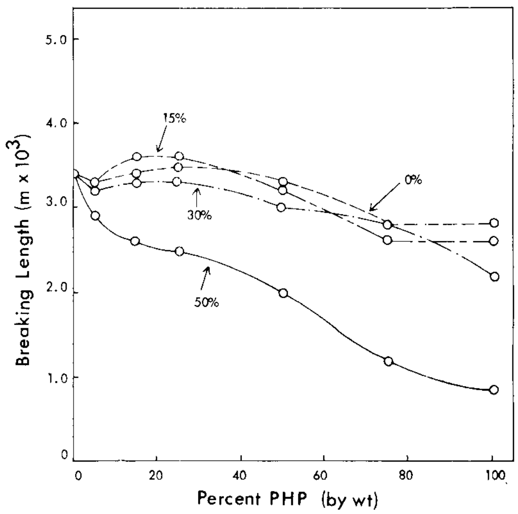

FIG. 4. Breaking Length of Blended Handsheets Using Bleached Peanut Hull Pulps.

ficient to lower the lignin content of the hull fibers to a permanganate number of around 22. The pulps obtained from the 0% and 15% sulfidity tests appeared to have been more completely delignified. The fibers were liberated to a greater extent; there were fewer undigested particles; and the products were of a lighter color when compared to the pulps received from the higher sulfidity tests. In general, the lignin content of the pulps appears to be lower at low sulfidity levels.

This unexpected observation of the effect of sulfidity content on delignification may find its explanation in a difference in lignin structure between peanut hulls and wood. Where sodium sulfide has been shown to effectively aid cleavage of glyceryl β-aryl ethers in the pulping of wood (Marton 1971), this ether linkage might not be as prevalent in peanut hulls as it is in the lignin of conifers and angiosperms. In contrast, phenolic hydroxyl groups might be predominating in the lignin of peanut hulls, causing it to dissolve more rapidly in alkaline solutions. Consequently, the concentration of sodium sulfide in the cooking liquor might be of less influence to the pulping of peanut hulls than it is to the delignification of wood. However, there seems to be some relationship between the sulfidity of the liquor and the shive content of the pulp with the effect that soda pulp has the least contamination from undelignified particles. At 0% and 15% sulfidity, the pulp apeared clean and homogeneous after bleaching with the shives entirely degraded. The higher sulfidity pulps seemed inferior.

Table 2 also shows the relation obtained between brightness and sulfidity level for the unbleached and partially bleached pulps. The data compare well with those typically obtained for wood pulps. For the unbleached pulps, the highest brightness was obtained at the lower sulfidity levels, although the brightness did not change markedly. For the bleached pulps, the brightness was independent of sulfidity up to 30%, beyond which the brightness decreased sharply.

Freeness, which is a measure of the drainage time of pulps, was determined for bleached and unbleached peanut hull pulps and results are contained in Table 2. For the unbleached pulps, the freenesses are relatively constant from 15% to 50% sulfidity and are independent of sulfidity. At 0% sulfidity, the freeness is slightly lower. For the bleached pulps, it is evident that the freeness is not increased by higher sulfidity. Freeness is a maximum at 0% sulfidity and decreases as sulfidity increases.

The results of strength tests with peanut hull paper and blended sheets are represented in Figs. 1–4. When compared to wood fiber paper, all of the hull papers were inferior. Handsheets formed from peanut hull pulp revealed that better paper was made at lower sulfidity levels than at higher levels. The soda-cooked pulps had the lowest content of shives among all pulps, and pulps from the cook with the highest sulfidity content had to be deshived prior to handsheet formation. Removal of these undelignified fiber bundles had a marked effect only on the breaking length of the 50% sulfidity peanut hull handsheets (Fig. 3), for which no good explanation can be given. Most other pulps contained considerably fewer shives for which reason screening was omitted altogether.

When tearing factors of blended handsheets were compared to those of handsheets made from softwood pulp (blotting paper), blended handsheets were inferior in quality if the content of peanut hull fiber exceeded approximately 20% as shown in Fig. 1. However, up to 100% strength increases over pure softwood pulp are observed if the handsheet contains only 5% unbleached peanut hull fibers. The increase in tear factor varies with the sulfidity level of the peanut hull cook and is highest at 15% sulfidity.

The results are similar, although less pronounced with bleached peanut hull pulp as shown in Fig. 2.

The breaking length of peanut hull paper also varies with the sulfide concentration of the cooking liquor. Blended handsheets with unbleached and partly bleached peanut hull pulps generally show slightly improved tensile strength properties over a wide range of blending when compared to paper made with pure wood pulp as is depicted in Figs. 3 and 4. Handsheets made with 5% peanut hull pulp of the 15% sulfidity cook show a striking strength increase of almost 50%. This value matches closely the observation of tear strength values.

CONCLUSIONS

It was found, for peanut hulls, that the degree of delignification was not distinctly different at different sulfidities and that the best results were obtained with liquors containing between 0–15% sulfidity. This suggests that the chemical nature of the lignin molecule contained in peanut hulls differs from that found in soft- and hardwood species.

Under the conditions outlined in this report, it was verified that a peanut hull pulp was of inferior strength to that of paper made of wood pulp. In contrast to results from previous investigations reported in the literature, however, yields of kraft pulped peanut hulls were found to range from 50 to 60%. This contradiction can probably best be explained with differences in cooking times, which were previously chosen to be up to 7 hr (Govil 1960). With respect to papermaking properties, it was found that when peanut hull pulp is added to wood pulp in proportions up to approximately 20% by weight, the strength of the paper product produced is comparable, and in several cases, increased over that of paper made with pure softwood pulp.

The tests showed that a conventional pulp and paper plant, designed for pulping wood, need not drastically modify its processes in order to utilize peanut hulls. Since the fibers are small, some problems could arise from using existing filtering apparatus, in which case a unique filtering system might be needed. However, in blends with wood fibers, this should not be a problem.

REFERENCES

Browning, B. L. 1963. The composition and chemical reactions of wood. Page 66 *in* B. L. Browning, ed. The chemistry of wood. Interscience Publishers, New York.

Chittenden, A. E., and E. R. Palmer. 1964. The production of particle board from groundnut shells, Board Practice 7(8):183–187.

Chittenden, A. E., and E. R. Palmer. 1965. Particle board from groundnut shells. Board Practice 8(3):75–82.

Govil, R. S. 1960. Chemical pulp from groundnut shells. Tappi 43(6):215A–216A.

Institute of Paper Chemistry. 1970. Pulping of bagasse and other papermaking fibers. Institute of Paper Chemistry, Appleton, Wisconsin.

JAIN, N. C., R. C. GUPTA, S. C. BAJAJ, AND D. D. SINGH. 1964. Utilization of groundnut shells. Ind. Pulp Paper 18(8):477–480.

LAI, Y. Z., AND K. V. SARKANEN. 1971. Isolation and structural studies. Pages 165–230 *in* K. V. Sarkanen and C. H. Ludwig, eds. Lignins—occurrence, formation, structure, and reactions. Wiley Interscience, New York.

LYNCH, D. F. J., AND M. J. GOSS. 1930. Peanut-hull cellulose. Ind. Eng. Chem. 22(8):903–907.

MARTON, J. 1971. Reactions in alkaline pulping. Pages 639–689 *in* K. V. Sarkanen and C. H. Ludwig, eds. Lignins—occurrence, formation, structure, and reactions. Wiley Interscience, New York.

MILLS, E. C. (ALSCOPE LTD.). 1969. Process for making pulp by impregnating cellulosic materials with ammonium hydroxide and nitric acid. U.S. Pat. 3,461,028.

MILLS, E. C. (ALSCOPE LTD.). 1970. Process for making pulp. Can. Pat. 854,073.

RYDHOLM, S. A. 1965. Pulping processes. Wiley Interscience, New York, pp. 677–714.