Optimizing alkaline pulping of wheat straw to produce stronger corrugating medium

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ABSTRACT: Corrugating medium is produced by carbonate–caustic (sodium carbonate–sodium hydroxide) pulping of hardwoods in U.S. mills. The method has never been used to produce corrugating medium from straw, a common fiber source in developing market economies. Cooking time and total chemical charge were varied in an attempt to optimize carbonate–caustic pulping of straw in terms of two important strength properties– Concora crush resistance and STFI short-span compressive strength. The optimized process requires a very low charge of caustic and provides strength values comparable with soda-pulped straw at a higher yield. The results imply that straw pulp mills, which typically operate without chemical recovery, could switch to carbonate–caustic pulping and build a carbonate recovery line at a fraction of the cost for a conventional recovery system.

KEYWORDS: Alkaline pulping, compression strength, Concora tests, corrugating medium, crush resistance, mechanical properties, optimization, short span tensile strength, soda pulping, wheat straw.

Straw was one of the first raw materials used for commercial production of pulp and paper. However, as a papermaking material, straw has several drawbacks compared with wood. Straw is difficult to collect, handle, bale, and store. Straw fibers also have low drainage characteristics. Finally, pulping generates a viscous black liquor with a low calorific value and a high silica content that renders the recovery operation difficult.

These technical and economic problems eliminated straw-pulp production in many developed countries. Many developing countries, however, do not have adequate wood supplies, and straw and other nonwood materials still constitute the major fiber source for pulp and paper production. According to Atchison (*1*), the worldwide average increase in production capacity for nonwood plant fiber is 4.7%, more than double the average 2% annual increase in wood pulp capacity. The greatest part of this increase is attributed to developing market economies, especially in Asia (*2*).

Most straw-pulp mills are relatively small, and economies of scale prevent them from building conventional chemical-recovery lines. This leads to higher production costs, inefficient use of resources, and environmental problems.

One of the major obstacles to chemical recovery—desilication of straw black liquors—has been overcome. There are two methods for desilicating black liquors. One uses milk of lime to precipitate silica as calcium silicates, which are removed by filtration. The new Naco process (*3*) uses a similar principle. The other desilication method uses flue gas to carbonate the black liquor and precipitate silica particles (*4, 5*). Now that the silica problem has been solved, recovery operations requiring less capital investment than conventional kraft recovery may be feasible for medium-sized straw-pulp mills.

Carbonate–caustic pulping

Corrugating medium has long been produced by carbonate–caustic (sodium carbonate–sodium hydroxide) pulping of hardwoods in U.S. pulp and paper mills. The method has never been used to produce corrugating me-

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dium from straw. Investigation of carbonate–caustic cooking of wheat straw is of practical interest because of the relative advantages of the chemicals in the recovery operation.

Sodium hydroxide is a more active chemical agent than sodium carbonate. Pulps prepared with sodium hydroxide are softer and easier to refine, resulting in a higher-strength pulp and lower energy costs. In carbonate–caustic pulping of wood, the optimum sodium hydroxide fraction is between 15–50% of the total chemical charge (as Na_2O), and the method has been patented by Owens–Illinois (*6, 7*). In carbonate–caustic pulping of wheat straw, the sodium hydroxide fraction in the cooking liquor is more important, since it determines the feasibility of the process. The need for a high sodium hydroxide fraction would make the pulping process nonfeasible, since conventional recovery of sodium hydroxide requires more investment dollars than the recovery of the sodium carbonate.

Study objectives

The aim of this study was to find the optimum sodium hydroxide fraction and cooking time to produce a straw pulp that provides a paperboard with maximum Concora crush resistance and STFI compressive strength. To evaluate the feasibility of the new pulping method, the maximum values for the two strength properties and the optimum cooking time must be compared with an existing pulping method. Thus, pulping conditions for soda (pure sodium hydroxide) pulping also were optimized. Soda pulping was chosen for comparison because it has been reported that kraft and NSSC (neutral sulfite semichemical) pulping do not have any advantages over soda pulping in terms of strength properties of straw pulps (*8*).

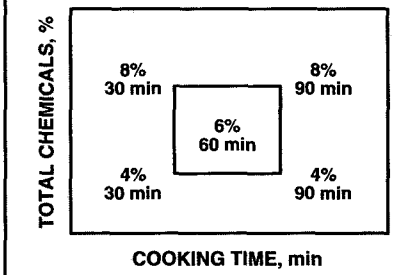
Optimization logic

For soda cooks, the optimization variables were chosen to be total chemical charge and cooking time at the maximum temperature. The optimum total

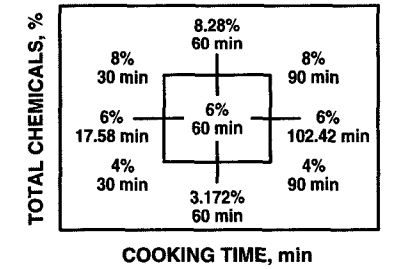
I. Soda cooking conditions and mechanical properties

Cook No.	Total chemical, %	Cooking time, min	Density, kg/m³	Refining time,s	Concora value, N	STC value, kN/m
1	6	60	550	33	286.14	4.10
2	8	30	550	30	283.30	4.10
3	4	30	452	90	245.60	3.86
4	6	60	550	38	290.59	4.16
5	4	90	550	64	278.21	4.54
6	8	90	550	25	271.05	3.95
7	6	60	550	48	288.63	4.10
8	6	60	550	39	280.22	4.02
9	8.828	60	550	29	271.67	3.94
10	3.172	60	391	90	179.38	3.00
11	6	17.58	550	90	318.31	4.29
12	6	102.42	550	52	285.73	4.00
13	4	30	452	90	270.78	3.67
14	8	30	550	43	296.90	4.00
15	5.5	17.58	550	72	319.64	4.07
16	7.0	17.58	550	60	308.03	4.10
17	6.5	17.58	550	90	329.30	4.15
18	6.25	17.58	550	48	308.87	4.19

1. Levels of the variables for the first-order experimental region (soda cooks)



2. Levels of the variables for the second-order experimental region (soda cooks)



II. ANOVA table for the first-order regression (soda cooks)

Source of variation	Sum of squares	Degrees of freedom	Mean square	F_0
Regression	336.805	2	168.403	0.74
Residual	...	5	...	...
Interaction	503.105	1	503.105	24.83[a]
Pure quadratic	568.182	1	568.182	28.04[a]
Pure error	60.789	3	20.263	...
Total	1468.881	7	...	...

[a]Significant at 5%

III. Carbonate–caustic cooking conditions and mechanical properties

Cook No.	NaOH fraction	Cooking time, min	Density, kg/m³	Refining time, s	Concora value, N	STFI value, kN/m
19	0.1	20	550	88	321.87	4.34
20	0.2	40	550	61	290.94	4.09
21	0.3	20	550	82	266.60	4.34
22	0.3	60	550	43	239.01	3.93
23	0.2	40	550	60	290.54	3.98
24	0.1	60	550	90	262.91	4.10
25	0.1	20	544	90	317.55	4.27
26	0.0	20	533	90	319.96	4.40
27	0.1707	34.144	550	62	296.73	4.11
28	0.1707	5.856	527	90	295.35	4.35
29	0.0293	5.856	477	90	262.24	4.11
30	0.0293	34.144	550	72	304.78	4.17
31	0.1	40	550	63	297.75	4.21
32	0.1	0	385	90	203.77	3.59
33	0.1	20	550	82	317.64	4.43
34	0.19707	20	550	75	315.02	4.29
35	0.1	20	550	86	315.68	4.40

chemical charge that was found as a result of these pulping experiments was fixed throughout the carbonate–caustic pulping experiments. The liquor-to-straw ratio (10:1) and cooking temperature (165°C) were also kept constant during both types of pulping experiments. These conditions were arrived at after a series of screening experiments.

During the optimization process, laboratory handsheets were compared at the same density. Refining time was used as an additional constraint, with a maximum refining time of 90 s in a PFI mill. If a particular cook failed to reach the required density (550 kg/m³) after 90 s, refining was stopped and lower-density handsheets were prepared and tested for crush resistance and compressive strength. This refining time was chosen because pulps from more than 80% of the experimental trials achieved the required density within 90 s.

Results

Soda cooks

A total of 18 cooks were performed in an effort to identify the optimum total chemical charge and cooking time with respect to Concora crush resistance. **Table I** lists the conditions applied during these cooks along with the corresponding Concora values. Optimum values for STFI compressive strength were also found within the same experimental region, and these values are included in Table I.

The first eight cooks were carried out to generate a first-order model around the center point of 6% chemical charge and 60 min, as illustrated in **Fig. 1**. The eight experiments were set so that they formed an orthogonal first-order design with four center points. The adequacy of the first-order model was checked by applying analysis of variance (ANOVA). Equation 1 is the first-order regression model that was found as a result of the first set of experiments.

$$\text{Concora (N)} = 244.88 + 3.82C + 0.17T \quad (1)$$

where

C = total chemical charge, %

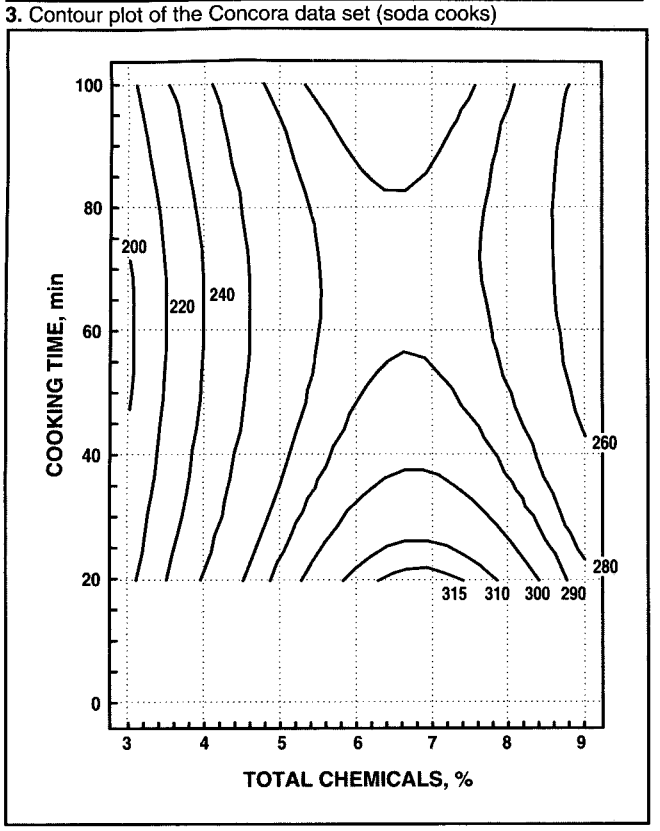
T = cooking time, min

Table II is the ANOVA table for the first-order regression. If the first-order model were found to be adequate, the center point would be moved to another location by applying the steepest-ascent method. At that point, another first-order model would be generated and its adequacy would be checked. The procedure would be continued as long as the first-order model was adequate. However, in this case, the ANOVA revealed that the first-order model was inadequate and that the surface had a curvature. Therefore, a second-order region was constructed around the same center point, as shown in **Fig. 2**, and cooks 9–14 were carried out.

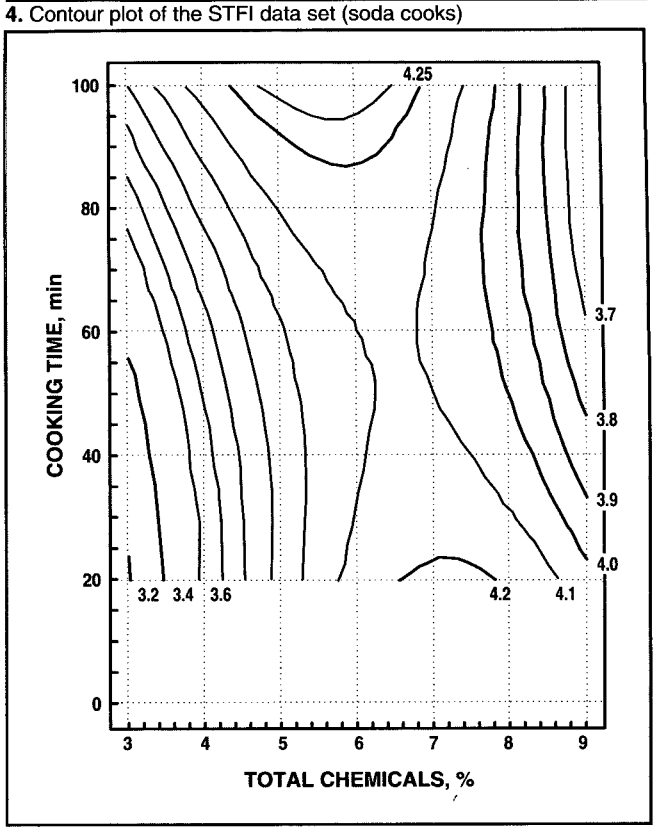
Response surface regression (RSREG) analysis (*9*) found the response surface as a saddle. Ridge analysis located the maximum response point around the 6.5% total chemical and 18-min cooking time at maximum temperature. Hence, four more cooks (cooks 15–18) at 5.5%, 6.25%, 6.5%, and 7.0% total chemical were made by fixing the time at 17.58 min to locate the maximum Concora value more precisely. **Figure 3** illustrates the Concora response surface in the form of a contour graph based on the results of the 18 cooks listed in Table I.

STFI was maximized using the same experimental region generated by the 18 soda cooks in Table I. **Figure 4** illustrates the STFI response surface within the experimental region. Ridge analysis for the STFI regression showed that the optimum chemical charge and cooking time were 5.7% and 102 min, respectively. However, the shape of the response surface for STFI is a saddle. Consequently, there is another steep edge with an STFI value close to the maximum that was found by the ridge analysis. Ridge analysis locates the maximum in the experimental borders of the study. However, a close investigation of the contour graph in Fig. 4 reveals another maxi-

3. Contour plot of the Concora data set (soda cooks)



4. Contour plot of the STFI data set (soda cooks)



mum point occurring at a much lower cooking time (20 min) and a higher chemical charge (7%). The Concora and STFI values at the optimum conditions were found to be 318 N and 4.31 kN/m, respectively, at a yield of 56.8%.

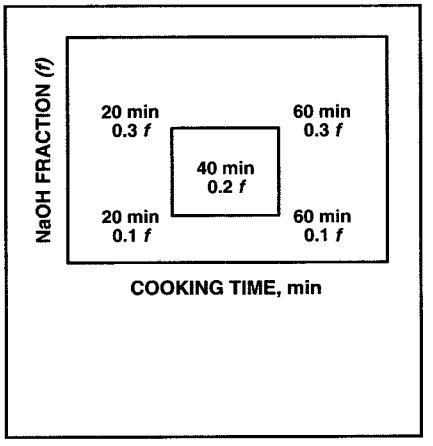
Carbonate–caustic cooks

A similar analysis was carried out for the carbonate–caustic cooks. A total of 17 cooks were carried out in an effort to maximize Concora crush resistance and STFI compressive strength. **Table III** lists the cooking conditions of the experimental design and the corresponding strength values.

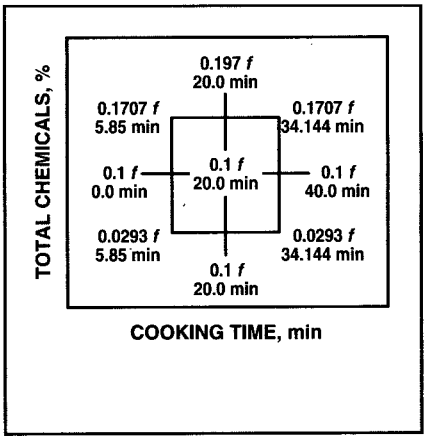
Six cooks (cooks 19–24), with two at the center point, were performed to generate the first-order model. The first-order region formed as a result of these cooks is illustrated in **Fig. 5**. Equation 2 is the first-order multivariate regression equation for this first set of experiments.

$$\text{Concora (N)} = 361.51 - 197.93f - 1.08T \quad (2)$$

5. Levels of the variables for the first-order experimental region (carbonate–caustic cooks). The *f* represents the NaOH fraction.



6. Levels of the variables for the second-order experimental region (carbonate–caustic cooks)



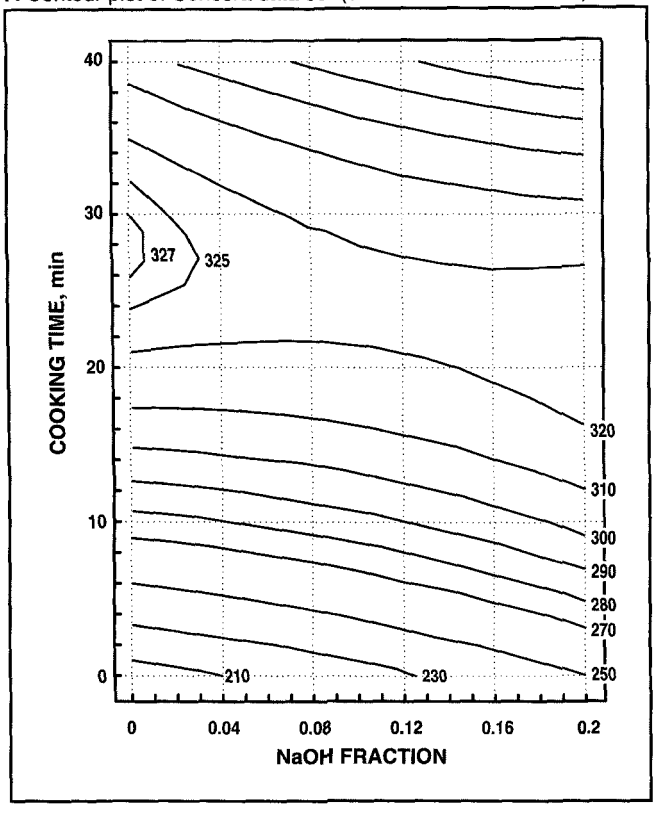
where

f = NaOH fraction

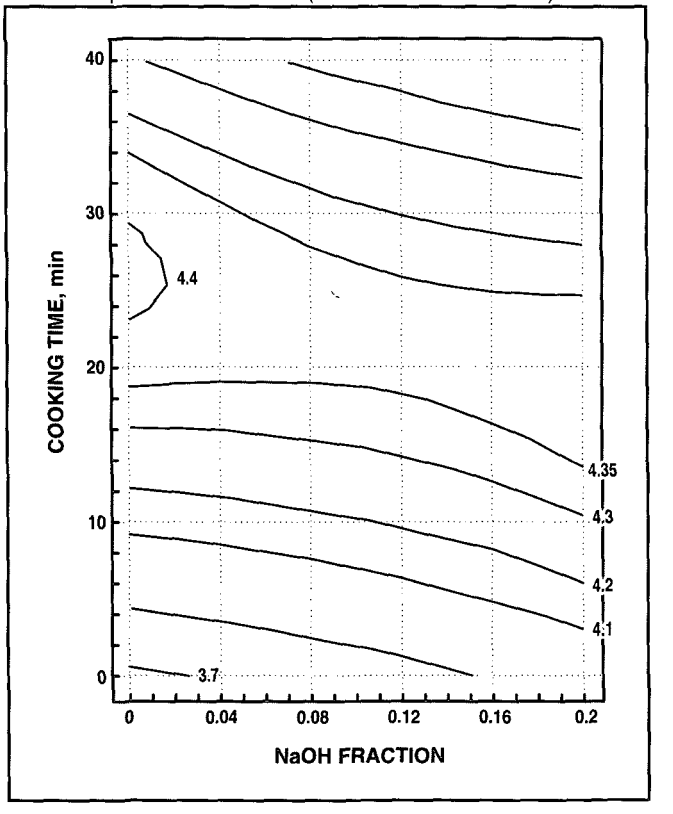
T = cooking time, min

Table IV is the ANOVA table for the first-order regression. Since the pure error was too small, all the terms (linear, quadratic, and interaction) were found to be significant. Therefore, the center point was shifted to the point where the maximum Concora value had been obtained after the first set of experiments (sodium hydroxide fraction = 0.1, cooking time = 20 min). **Figure 6** shows the second-order region constructed around the new center point for the second stage of the carbonate–caustic experiments (cooks 25–35). The design recognizes the fact that negative sodium hydroxide frac-

7. Contour plot of Concora data set (carbonate–caustic cooks)



8. Contour plot of STFI data set (carbonate–caustic cooks)



tion and negative cooking time are inadmissible. The RSREG (*9*) analysis determined the Concora response surface as a maximum when all 17 data points (cooks 19–35) were taken into account. When the second-order region alone was considered, the response surface was a saddle. **Figure 7** illustrates the contour plot of the surface for the Concora data set.

Figure 8 represents the STFI surface found in the same region. The optimum sodium hydroxide fraction for the maximization of both strength properties were found very close to zero—0.007 for the Concora and 0.004 for the STFI. Ridge analysis located the optimum cooking time at the maximum temperature for Concora and STFI as 27 min and 25 min, respectively. The maximum Concora and STFI values were found to be 327 N and 4.41 kN/m, respectively, at a yield of 62%.

Figures 3 and 4 show that the optimum cooking time for soda cooks is in the range of 20 min. For the caustic–carbonate cooks, the optimum time is in the range of 20–30 min, depending on the sodium hydroxide fractions used. The higher the fraction, the lower is the cooking time needed to achieve a specific level of strength property.

Experimental

Raw materials

A bale of the main raw material—soft, white winter wheat straw (*Triticum aestivum*)—was purchased in the Kalamazoo area. The straw was cut to lengths of 5–7.5-cm with a guillotine cutter and was hand-cleaned prior to pulping. A long-fibered basestock was mixed with the straw pulp. The basestock was obtained in the form of grocery bags. These were repulped and mixed with the straw pulps to the extent of 25% of the total stock. Sheets prepared from 100% grocery bag pulp had lower strength properties than the sheets prepared from the mixture of straw pulp and basestock. The Concora and STFI values obtained at the same basis weight were 213 N and 3.4 kN/m, respectively.

Cooking and washing

A 176-g sample of oven-dry (o.d.) straw was cooked in an M&K digester with a liquor-to-straw ratio of 10:1 and a preheating time of 35± 3 min. The cooked straw was washed in running tap water, centrifuged, and then disintegrated in a Waring blender at 1.5% consistency. This pulp was filtered, washed, and centrifuged again and stored in dark bags in a refrigerator at 4°C. The kraft bags were shredded and soaked in process water for 12 h prior to defiberizing in a Valley beater at 1.57% consistency for 1 h. The pulp was then screw pressed, centrifuged, and stored in dark bags at 4°C. The kraft pulp thus obtained had a freeness of 450 mL CSF.

Refining

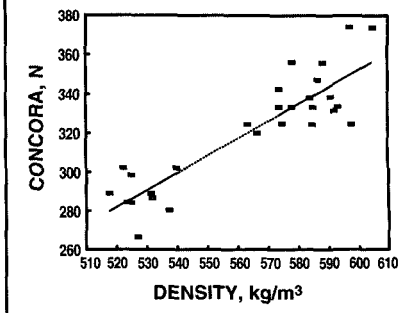
The straw pulps were refined in a PFI mill at 10% consistency in 21-g batches for varying times to obtain different sheet densities. Final strength properties were compared at a constant density of 550 kg/m^3 or at a refining time of

IV. ANOVA table for the first-order regression (carbonate–caustic cooks)

Source of variation	Sum of squares	Degrees of freedom	Mean square	F_0
Regression	3439.70	2	1719.85	7.53[a]
Residual	...	3	...	...
Interaction	246.02	1	246.02	3068.06[b]
Pure quadratic	438.75	1	438.75	5473.58[c]
Pure error	0.08	1	0.08	...
Total	4124.55	5	...	...

[a]Significant at 10%
[b]Significant at 5%
[c]Significant at 1%

9. Concora regression line for cook no. 28



90 s, whichever was achieved earlier.

Sheet formation

The pulp from kraft bags was disintegrated in a standard disintegrator at 0.7% consistency for 15,625 revolutions (5 min). The pulp was then mixed with refined straw pulp in the ratio of 25:75 in the homogenizer for 10 min at 0.14% consistency. Handsheets from this homogenized slurry were prepared in a Noble & Wood machine to provide sheets of 5.25± 0.05 g (127 g/m^2, or 26-lb sheets). The handsheets were stored in a preconditioning room held at 35% RH and 72°F for 24 h and then for 2 h in a conditioning room held at 50% RH and 72°F.

Determination of strength properties

For the Concora crush-resistance tests, ten 152-mm x 12.7-mm (6-in. x 0.5-in.) strips were cut from each handsheet using a Concora diecutter. For the STFI compressive-strength tests, five 177-mm x 15-mm (7-in. x 0.6-in.) strips were cut from the remaining portion of each handsheet using a TMI precision cutter. Density was determined by weighing and measuring the thickness of each sample. The strips were tested for Concora crush resistance and STFI compressive strength according to TAPPI T 809 om-87 and T 826 pm-86, respectively. For each cook, two simple linear regression lines, one for the Concora crush resistance and one for the STFI compressive strength, were fitted against sheet density. **Figure 9** shows a typical regression line for Concora crush resistance. The strength values corresponding to density of 550 kg/m^3 were calculated from these regression equations and used to generate data sets for statistical analysis.

Statistical design of the experiments

The statistical design used in the experiments was one of response surface methodology (RSM). This approach was chosen because we deemed it more efficient than either a conventional factorial design or the Taguchi method of orthogonal arrays. The design involves construction of a mathematical model to locate the optimum if it exists within the experimental region. If it does not, the experiments are continued along a gradient of steepest ascent in an effort to locate the center of a new experimental region to be tested. Another round of experiments are conducted around this new point to determine whether the new region contains the optimum. For the traditional factorial design methods or the Taguchi methods to be equally successful, a correct choice of experimental conditions must be known before beginning the experiments. Those methods are comparatively more sensitive to the choice of levels for each factor as well as the range for each factor. A more in-depth study of the different methods can be found in a standard text (*10*).

Conclusion

The results of this study show that during carbonate–caustic pulping of wheat straw, a very low amount of sodium hydroxide (less than 0.01 fraction of the total chemical charge) is enough to provide the same strength values that are obtained with soda pulping. Carbonate–caustic pulping also provides pulp at a higher yield.

However, cooks containing higher sodium hydroxide fractions are easier to refine. At the level of refining energy chosen for use in this study, the optimum sodium hydroxide fraction works out to be 0.195. If the mill can tolerate a higher level of refining energy, this fraction can be lowered. Thus the precise amount of caustic to be used in a given pulp mill depends on local economic conditions. The data also suggest that cooking times would be longer for liquors containing low caustic fractions, which could result in greater carbohydrate dissolution (*11*).

A straw pulp mill that adapted to carbonate–caustic pulping could build a carbonate chemical recovery line for less cost than for a conventional chemical recovery operation. ▮

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