RECYCLABILITY OF MIXED OFFICE WASTE PAPERS CONTAINING PRESSURE SENSITIVE ADHESIVES AND SILICONE RELEASE LINERS

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Abstract
Increased use of pressure sensitive adhesives for labels and stamps has introduced another contaminant into the office paper stream: silicone-coated release liners. This study examines methods and conditions for removal of contaminants, including these liners, from a typical batch of discarded office papers. Removal of contaminants contained in the furnish were compared using alkaline and near neutral pH repulping conditions. Residual contaminants were measured by image analysis, and the number and size of the contaminants were compared on handsheets that were prepared from samples collected at stages from pulping through bleaching. Handsheets were also tested for tear and tensile strength. Results showed that unadjusted pH 8.6 was more effective than pH 10.0 for contaminant removal. Flotation was the most effective stage for removal of contaminants. Presence of residual adhesives or silicone did not compromise final paper strength properties to a great extent; pH 10.0 repulping produced only slightly higher tensile and tear properties than did pH 8.6 pulping.

Introduction
An increased use of pressure sensitive labels and stamps has increased the level of potential contaminants such as silicone release liner and adhesives in the mixed office waste material stream. The presence of liner may increase the total number of contaminants, which eventually leads to problems in the recycled paper quality and process equipment performance [1]. As a result, it is important to understand how silicone release liner interacts with pressure sensitive adhesives (PSAs) and inks present in mixed office papers. Little research has been reported on this topic.

Currently, there are few ways to improve the quality of the recycled fibers containing these contaminants or to decrease the disposal of large volumes of waste material. One attempt involved a process that recycles silicone-coated paper through pulping in the presence of a salt of monophosphoric acid ester [2]. Also, the United States Postal Service (USPS) is in the process of evaluating adhesives and liners to assure products used on their stamps and labels are recyclable [3]. However, there are numerous PSAs and liners on the market.

Our study used mixed commercially available labels and liner materials at maximum levels that would be found in a typical office paper mix. Continuing research with this mixed office waste is important to develop ways to utilize this material and to prevent major contamination of recycled pulp.

Experimental
A large batch of laser-printed white office copy paper collected at the USDA Forest Service, Forest Products Laboratory (FPL), was shredded for this study. Pressure sensitive adhesive labels and their corresponding silicone release liners were primarily preconsumer stock from the FPL stationery supplies.

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Address labels received in the mail from various sources comprised the remainder of the mix. Labels were applied to the copy paper. Six 3-kg runs were pulped, screened, floated, and bleached. The composition of the runs was as follows:

Run 1. 100% printed copy paper (control)
Run 2. 90% printed copy paper and 10% silicone release liners
Run 3. 85% printed copy paper, 10% release liners, and 5% adhesive labels
Run 4. 95% printed copy paper and 5% adhesive labels

The pH of these runs was adjusted to pH 10 in the pulper with 5% sodium hydroxide. Runs 5 and 6 were performed with an unadjusted pH of 8.6 contributed by calcium carbonate fillers and city-softened water.

Run 5. Same as Run 3
Run 6. Same as Run 3, plus 0.025% amylase and 0.025% cellulase (based on ovendry weight of paper) Enzyme preparations, Termamyl 300L and Novazym 613, were provided by Novo Nordisk Biochem of North America, Inc. (Franklinton, NC)

The batches were pulped at 12% consistency for 25 min at 45°C in a 50-kg hydrapulper, screened through a vibrating 0.006-in. (0.15 mm) slotted screen at 45°C, and floated in a 80-L flotation cell. Flotation was carried out at 1% consistency with a 0.1% nonionic surfactant (BRD 2340 from Buckman Laboratories, Inc., Memphis, TN) for 5 min and was followed by washing. Handsheets were prepared from the pulper, screen accepts, and flotation accepts. Ovendry 50-g samples of the flotation accepts from each run were bleached with hydrogen peroxide and formamidine sulfinic acid (FAS) for additional handsheet analysis. Peroxide bleaching involved samples at 14% consistency with 1% peroxide, 0.75% sodium hydroxide, 3% sodium silicate, and 0.05% magnesium sulfate. The sample was heated to 80°C and put in a water bath for 90 min. The FAS bleaching was at 4% consistency and was performed with 0.5% FAS and 0.25% sodium hydroxide. The samples were heated to 60°C to 65°C and put in a water bath for 45 min.

Residual contaminants on handsheets from all sampling points were counted with the Apogee Spec Scan system (Apogee Systems, Inc., Powder Springs, GA) using a HP Scanjet 6100C flatbed scanner (Hewlett-Packard, Palo Alto, CA). Particles were counted with a size range down to 0.02 mm^2. Handsheets were recounted after being dyed according to the USPS protocol used at FPL [4]. The handsheets were dipped in the hydrophilic Morplas Blue 1003 dye solution (Pylam Products Co., Inc., Tempe, AZ) to stain the colorless adhesive and silicone particles a dark blue. Heptane was used to solubilize the dye. The dyed contaminant particles and toners are easier to see in contrast with the light background. Tensile and tear properties were measured from each run according to TAPPI T 994 om-96 for tensile and TAPPI 414 om-98 for tear.

Results and Discussion

Figures 1 and 2 show the summarized results for the average parts per million of residual contaminants from the image analysis of the undyed and dyed handsheets. Forward and reverse cleaners that separate contaminants by density were not used in this study. Although the valley screen separates large particles, screening on a larger scale involves multiple pressure screens and often multiple flotation stages. The results reported here show only the relative contaminant removal using valley screening and single flotation.

We found that flotation was the most effective process stage for removing both toner inks and the silicone. Tables 1 to 3 contain a summary of contaminants detected on undyed and dyed handsheets at each processing stage. For example, undyed handsheets for Run 2 had an average of 3,650 ppm of contaminants after pulping. This value dropped to 455 ppm after flotation. The dyed handsheets for Run 2 had an average of 11,396 ppm after pulping, which dropped to 901 ppm after flotation. Adhesives proved to be the most difficult contaminant to remove and were taken out with screening. Undyed handsheets for Run 4 had an average of 11,028 ppm after pulping. Once the pulp was screened, this value dropped to 6,410 ppm. Dyed handsheets for Run 4 had a value of 36,166 ppm after pulping, which dropped to 28,416 ppm after screening. Because the adhesives collected toner particles during pulping, these particles were visible in the undyed sheets as well as the dyed sheets. There was a large number of counts per square meter as well as a small average particle size (0.04 mm^2) in the pulper with the silicone liner (Run 2; Tables 2 and 3). In Run 3, with the addition of adhesive, particle size was 0.14 mm^2 (Run 3). The same mix run at neutral pH (Run 5) averaged a particle size of 0.19 mm^2 (Table 3).

Recycling mills repulp at either neutral or alkaline pH, so we compared the influence of pH on this furnish. The most significant observation was the impact of pulping pH on subsequent contaminant removal. Run 3 at pH 10.0 had a flotation value of 833 ppm for the undyed handsheets and 2,794 ppm for the dyed. At the neutral pH, Run 5 had a floated value of 748 ppm undyed and 2,043 ppm for dyed handsheets while Run 6 had a value of 437 ppm undyed and 1,603 ppm dyed. Neutral pH pulping is a gentler process than alkaline pulping and permitted more efficient removal of the adhesive. Toner particles were collected by the adhesive during pulping, which made many of the adhesive particles visible, and they were therefore counted on the undyed as well as the dyed handsheets. The gentler pulping keeps the contaminant particles large, whereas alkaline pulping solubilizes or fragments the silicone and adhesive.

Bleaching and washing also improved contaminant removal. The high alkalinity and temperature used for hydrogen peroxide bleaching fragmented residual contaminants permitting these particles to be washed from the pulp slurry. At pH 8.6, Run 5 had a flotation value of 748 ppm undyed and 1,354 dyed. When the pulp was peroxide bleached, this count was lowered to 421 ppm undyed and 1,277 ppm dyed. Reductive bleaching with FAS was less effective than the oxidative peroxide bleaching for reducing residual contaminants. Run 5 only dropped to 709 ppm undyed and actually increased to 2,219 ppm when dyed. However, the standard deviation on that point was large.

Table 4 presents the tensile and tear strength data. There was little difference in tensile strength values between the runs. At pH 10.0, Run 3 had a value of 34.9 Nm^2/g, and at pH 8.6, Run 5 had a value of 34.7 Nm^2/g. While contaminants decreased tensile slightly, the largest decrease occurred in Run 6 with a value of 31.4 Nm^2/g. Usually enzyme deinking has no negative impact on strength properties at low levels and short retention. We cannot explain this reduced strength.

Tear strength analysis also showed little difference between the different runs. Run 2 at the alkaline pH had a value of 8.8 kNm/g, and Run 5 at the neutral pH had a value of 8.7 kNm/g. The largest decrease again came with Run 6 with a tear value of 8.4 kNm/g. Despite this decrease, it can be concluded that the pH adjustment between the alkaline and neutral trials did not significantly affect the strength of the handsheets.

Deinking of toner-printed paper can be improved by the application of enzyme. The amylase–cellulase combination is especially effective as reported by Gehlhoff [5] and Zollner [6]. The most contaminant removal occurred with the use of the enzyme mixture in Run 6. At pulping, Run 6 had an undyed handsheet average of 6,089 ppm and a dyed handsheet average of 24,551 ppm. These counts dropped to 242 ppm undyed and 648 ppm dyed after peroxide bleaching.

Conclusion

We found that the best method for removal of silicone and pressure sensitive adhesive were those runs at pH 8.6. The flotation stage was the most effective at removing silicon, and screening was the most effective for removing adhesives. Of the pH 8.6 trials, the run with the enzyme (amylase–cellulase) provided the most contaminant removal. The tear and tensile strengths of these runs were not substantially affected by the adjusted pH of pulping or the residual contaminants. This study suggests that up to 10% silicone-coated release liners could be satisfactorily recycled in mixed papers.

Future Plans

Based on the results of this study, we plan to explore enzymatic removal of contaminants more completely.

References

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Table 1: Average parts per million of contaminants detected on undyed and dyed handsheets at each processing stage

	Average ppm									
							Bleaching			
	Pulper		Valley screen		Flotation		H_2O_2		FAS[a]	
Run	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed
1	3,967	10,211	4,021	8,940	528	3,338	287	963	504	1,177
2	3,651	11,396	3,483	8,245	455	901	238	865	403	1,679
3	10892	30,835	4575	17,203	833	2,794	585	1,760	702	1,981
4	11,028	36,166	6,410	28,416	1,560	6,779	855	4541	1,355	5,349
5	8,402	31,465	4868	13,793	748	2,043	421	1,277	709	2,219
6	6,089	24,551	4,103	10,180	437	1,603	242	648	310	1,641

[a]FAS, formamidine sulfinic acid.

Table 2: Counts per square meter of contaminants detected on undyed and dyed handsheets at each processing stage

	Counts/m^2									
							Bleaching			
	Pulper		Valley screen		Flotation		H_2O_2		FAS[a]	
Run	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed
1	87,174	217,633	85,574	187,278	10,970	54,749	6,755	21,163	11,026	24,068
2	83,215	245,197	80,053	283,880	8,527	16,833	5,389	18,618	7,704	29,743
3	109,642	218,884	91,740	250,496	10,018	21,158	6,759	12,588	8,849	16,500
4	110,707	257,058	116,355	332,435	19,111	48,863	9,778	26,990	17,502	40,958
5	78,865	161,301	99,700	229,831	12,847	26,932	7,483	17,061	11,658	27,830
6	74,411	237,017	77,660	159,790	8,298	22,818	4,624	9,675	6,056	23,947

[a]FAS, formamidine sulfinic acid.

Table 3: Average particle size of contaminants detected on undyed and dyed handsheets at each processing stage

	Average particle size (mm^2)									
							Bleaching			
	Pulper		Valley screen		Flotation		H_2O_2		FAS[a]	
Run	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed	Undyed	Dyed
1	0.0455	0.0469	0.0470	0.0477	0.0481	0.0610	0.0425	0.0455	0.0457	0.0489
2	0.0439	0.0465	0.0435	0.0290	0.0534	0.0535	0.0441	0.0465	0.0523	0.0565
3	0.0993	0.1409	0.0499	0.0687	0.0832	0.1321	0.0866	0.1398	0.0794	0.1200
4	0.0996	0.1407	0.0551	0.0855	0.0817	0.1387	0.0874	0.1683	0.0774	0.1229
5	0.1065	0.1951	0.0488	0.0600	0.0582	0.0759	0.0563	0.0748	0.0608	0.0798
6	0.0818	0.1036	0.0528	0.0637	0.0527	0.0703	0.0524	0.0669	0.0512	0.0685

[a]FAS, formamidine sulfinic acid.

Table 4: Comparisons of tear and tensile strength

Run	Tear index (kN*m/g)	Tensile index ($N*m^2/g$)
1	9.0	36.1
2	8.8	34.0
3	8.8	34.9
4	8.6	31.9
5	8.7	34.7
6	8.4	31.4

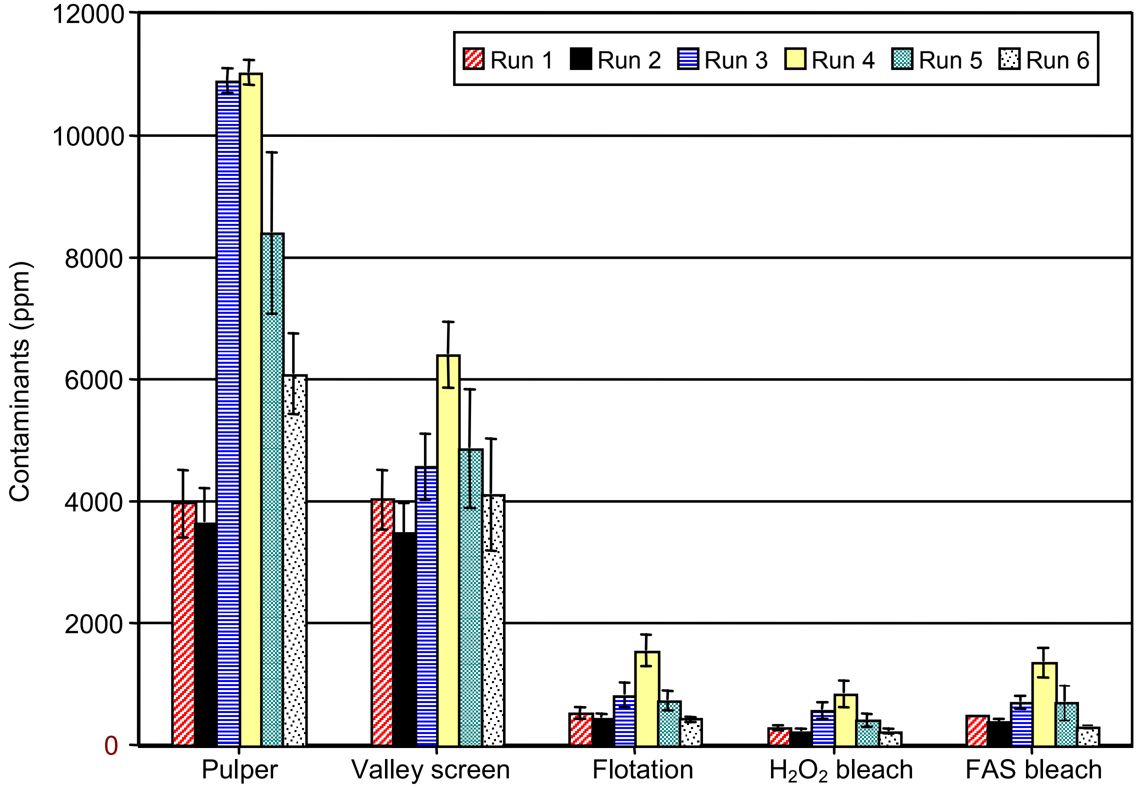

Fig 1: Residual contaminants measured by image analysis on undyed handsheets at each processing stage (FAS, formamidine sulfinic acid).

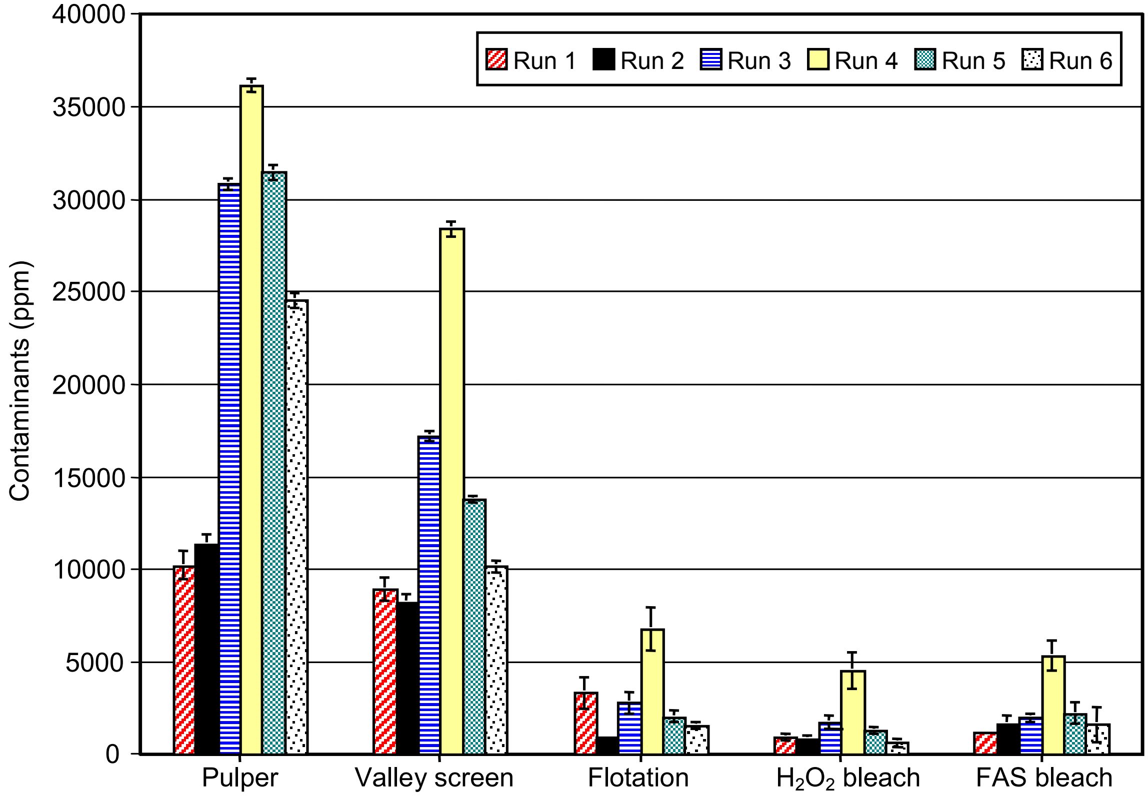

Fig 2: Residual contaminants measured by image analysis on dyed handsheets at each processing stage (FAS, formamidine sulfinic acid).

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