

Deinking laser-print paper using ultrasound

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ABSTRACT: *Laser-printed pulp is distinguished by large, flat, platelike ink particles. These particles are much more difficult to remove than conventional ink particles. It was shown in this research study that ultrasound waves can split apart and loosen these tenacious particles. Use of a particle-size analysis system suggested that frequencies of 22 kHz, 34 kHz, and 54 kHz each affected particles of different size ranges. This implies that a commercially feasible ultrasound deinking process might be developed as a stand-alone treatment method for laser-print papers or as a polishing step for mixed post-consumer wastes to improve the overall pulp quality.*

KEYWORDS: *Deinking, lasers, reclaimed fibers, ultrasonic frequencies, waste papers.*

One of the rapidly growing areas of the pulping industry is the use of secondary fibers. In 1990, the United States recovered about 33.3% of its post-consumer wastes, or 28.9 million tons, up from about 23% in 1988. The goal of the U.S. paper industry is to increase that percentage to at least 40%.

The contaminants which must be removed prior to fiber reuse vary with the grade of the paper and the intended end use. They include hot melts, latexes, and inks. The higher grade papers, such as newsprint and some office forms, primarily have to be deinked before reuse.

Though the "traditional" deinking processes used commercially are proprietary, all are essentially cleaning

processes: a combination of chemicals and mechanical forces are used to dislodge the ink particles from the fibers and disperse them in an aqueous medium. The ink particles are then separated from the stock by washing or by flotation or by both methods. The primary chemicals used for deinking are surfactants, which are chemicals that modify the surface properties of liquids and solids. Specifically, detergents, dispersants, and foaming agents are required. Generally, these are chemically modified mineral oils. Cautic soda, sodium silicate, and borax may also be added to enhance the surfactant action.

A limitation of all of these conventional deinking processes is their inability to effectively deink laser-printed

paper. This segment of the recycle market is rapidly growing. In many cases, the laser-print paper is not separated from the other office papers. The use of such mixed grades can lead to a lower quality sheet. At the same time, recent reports and articles (1) have confirmed the desire of consumers for a cleaner recycled sheet. Just because it is "recycled" is not the magic needed to maintain market share!

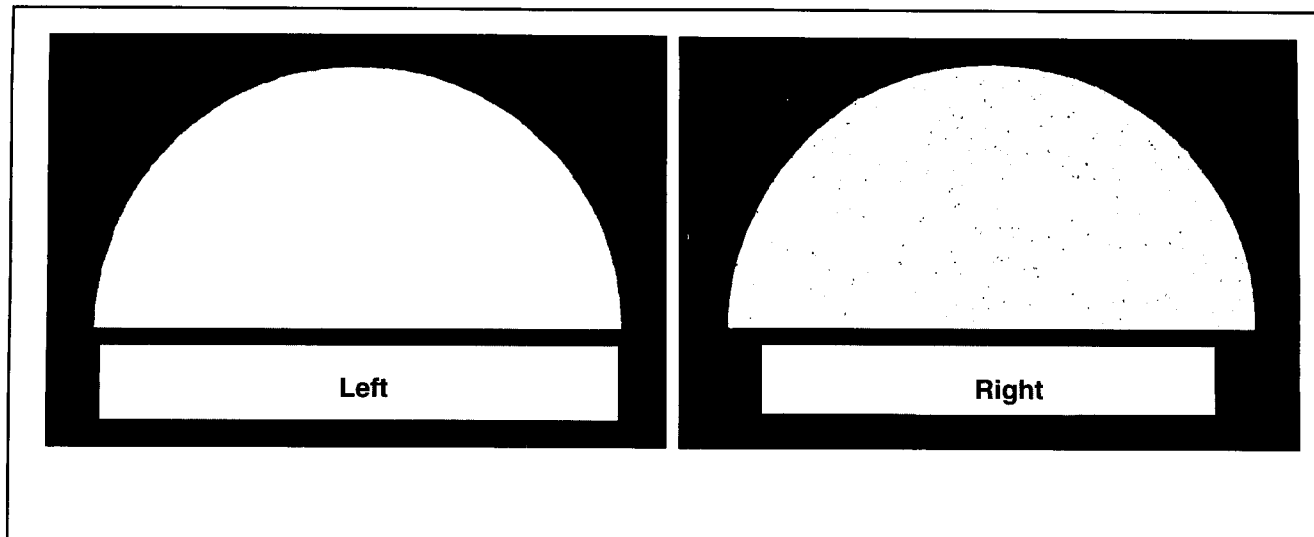
Recently a number of new processes have been developed for use on laser pulps. For example, Betz PaperChem markets a process which agglomerates rather than disperses the ink particles (2). A similar process was reported by Darlington of PPG Industries in 1989 (3). These agglomeration processes, though based on different physical principles, nonetheless require chemical treatment as do the more conventional deinking techniques. Other researchers have suggested using high-consistency pulping followed by flotation (4), the use of slotted pressure screens (5), or the adsorption on polystyrene foam prills (small, beadlike pellets) added to the pulper (6).

Another possibility for deinking both laser and ordinary high-grade office papers is by the use of ultrasound waves. Ultrasound waves are those sound waves with frequencies too high to be heard by the human ear, or greater than about 16 kHz. The use of sound waves to deink secondary fibers has been studied by two research groups in the United States—Turai and Teng (7) at SUNY-Syracuse and Naimpally (8) at California State University-Long Beach. Both research

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1. Handsheets prepared from ordinary pulp (left) compared with handsheets prepared from a 100% laser furnish (right).



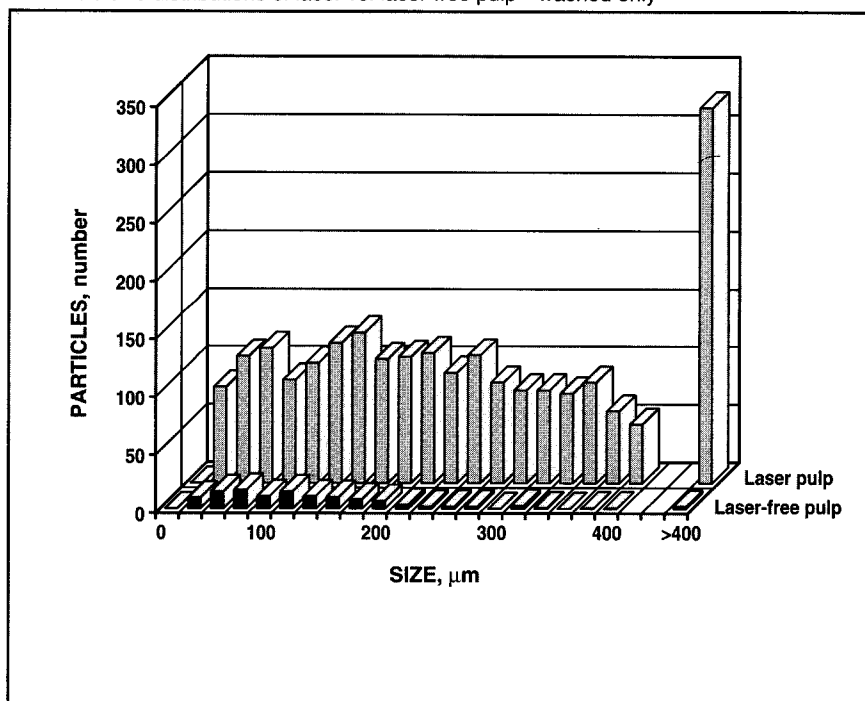
groups used a sonolator, a liquid whistle ultrasound generator producing waves at 22.5 kHz. These tests were very limited and did not include the study of laser-printed stock, but the results showed much promise both for deinking and for paper property development (tensile, burst, stretch).

The purpose of this study was to investigate the feasibility of deinking laser-printed secondary fibers using electromechanically produced ultrasound. The use of either magnetostrictive or piezoelectric crystals to generate the ultrasound waves eliminated the possibility that some of the cleaning action was due to mechanical action induced by the experimental setup, not the ultrasound waves themselves, as might have been the case when using a liquid whistle. It was hoped that the ultrasound treatment would also increase the fibrillation of the fibers, thus lead to an increase in paper strength. In addition, several different frequencies (22 kHz, 34 kHz, and 54 kHz) were used so their effects on different sized ink particles could be monitored.

The nature of laser-printed paper

Originally the term "laser print" meant specifically the computer-generated printing performed on a printer that uses a scanning laser beam as its light

2. Particle size distributions of laser vs. laser-free pulp—washed only

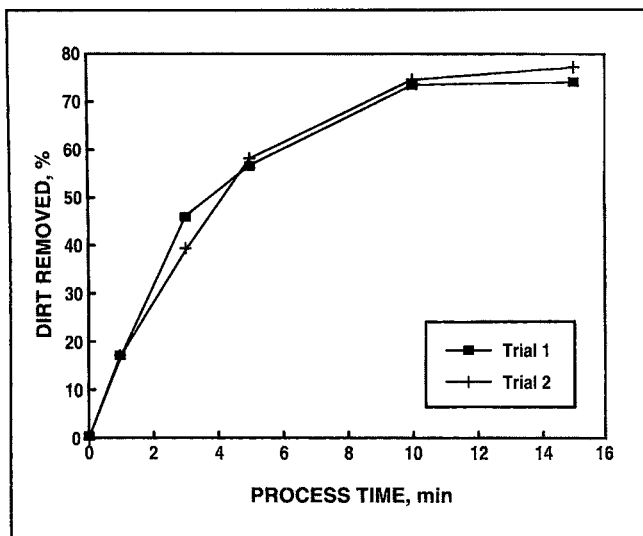


source. In recycle jargon, the expression has recently come to include all material which has been printed by using a "dry ink" or "toner" and a heat fixing process to coalesce the toner particles and attach them to the paper. Consequently, xerographic papers are commonly included in this classification (9).

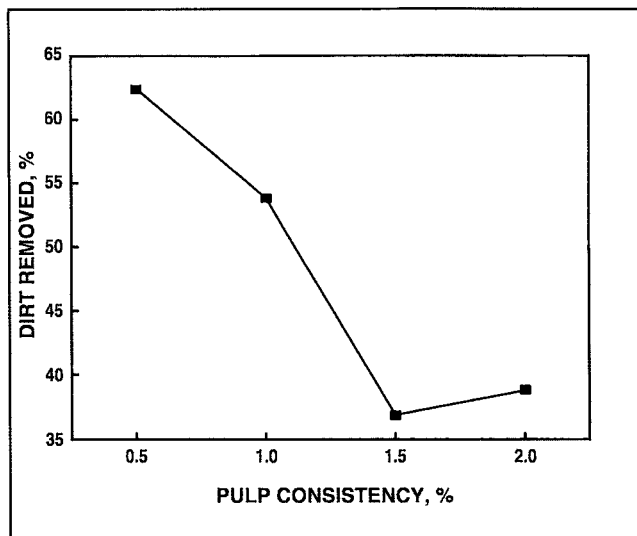
Xerographic inks are different from

actual laser-printed inks. Dry process xerography inks are thermoplastic resins (such as copolymers of styrene and acrylate) mixed with a pigment, generally carbon black. The materials are formulated so they become tacky at 70–120°C. Though they are not very susceptible to chemical treatment, mechanical forces can be used to disperse the ink into fine particles. Consequently, a combination of washing

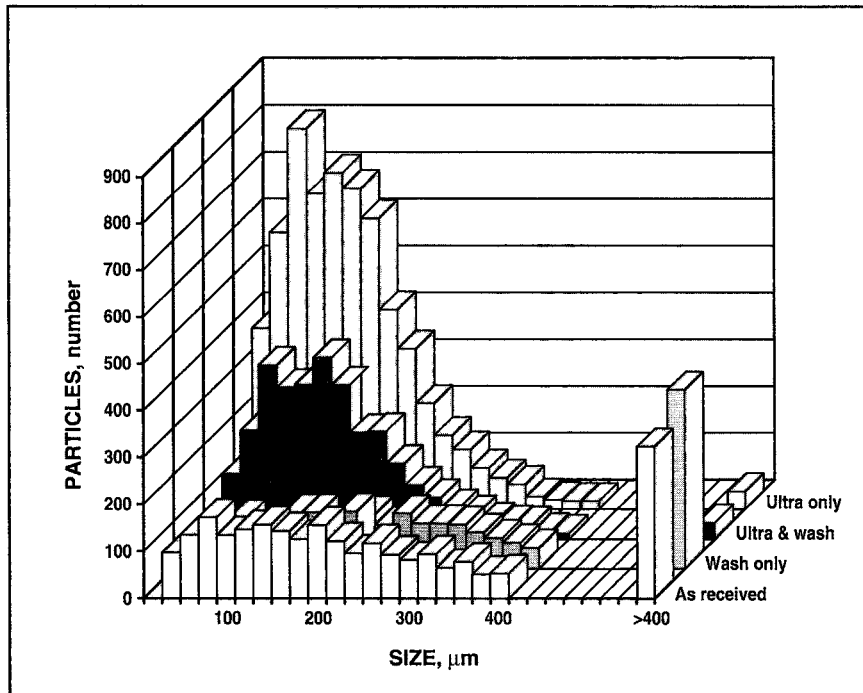
3. Percent cleanup vs. time



4. Consistency vs. dirt removal



5. Separation of process steps



and flotation has been shown effective in removing these inks (4).

True laser ink particles, on the other hand, are flat and plate-like, and contain tough binders (such as polyethylene and polypropylene) that are not easily reduced in size. Washing, therefore, is not effective in removing these inks (4). Because of this difficulty in deinking these fibers, we chose to use a 100% traditional laser furnish, pre-

pared directly from the output of a laser printer, in this study. The sheets that were pulped were very densely printed, giving a "worst case" scenario.

Figure 1 compares handsheets prepared from ordinary pulp and from a 100% laser furnish. As can be seen, the laser pulp has many more large ink particles.

Figure 2, which compares the par-

ticle size distributions of the two types of pulp, quantifies this difference.

Mechanism of ultrasound cleaning

When a sound wave traverses a material, the molecules of the material are induced to vibrate longitudinally in the direction of wave propagation. This results in pressure variations, which in a liquid lead to cavitation or the formation of tiny bubbles about 0.1–0.2 mm in diameter.

A bubble in a liquid is inherently unstable. Usually large bubbles will float away and burst on the surface and very small bubbles will redissolve in the liquid. When, however, the bubble is irradiated with ultrasound, it can continually absorb energy from the alternating compression and expansion cycles of the sound wave. This can lead to either a bubble that simply oscillates in size or to one whose average diameter will increase.

As a bubble grows, it will eventually reach a critical size where it will most efficiently absorb energy from the ultrasound. At this point, it will first grow very rapidly, and then collapse. As these bubbles collapse, or implode, violently in less than a microsecond, the temperature in their interior can reach 5500°C. Because the volume is so small,

this heat dissipates very quickly. At any given time, the bulk liquid remains near the ambient temperature.

The critical size depends on the frequency of the ultrasound. At 20 kHz, for example, the critical size corresponds to a bubble about 0.17 mm in diameter (10).

The imploding bubbles can transmit energy to the nearby medium. In the case of secondary fibers, the energy from these imploding bubbles can have two major effects: it can loosen the ink particles, removing them from the fibers, and it can split apart the ink particles themselves.

The speed of sound varies with the medium, increasing with increasing density of the medium. In water, for example, it is 4708 ft/s (1435 m/s); in wood (oak, with the grain) it is 12,620 ft/s (3850 m/s). It would be expected that the speed in the pulp slurry is intermediate between these two values. At 20 kHz, therefore, the wavelength is on the order of 4 in. (10 cm).

Experimental

Equipment and general procedures

The majority of our tests were made using a Fischer Scientific Model 550 Sonic Dismembrator. The Dismembrator is a self-contained unit which generates 20-kHz ultrasound waves. These waves are transferred to the pulp slurry by a probe which can be inserted into a container of pulp. The maximum power rating on the Dismembrator is 475 W; however, during normal operation, the efficiency rating (as read on the front panel) was typically 39%, or about 185 W.

To compare the effect of different frequencies, a series of ultrasonic transducers was mounted with epoxy directly on the inside bottoms of stainless steel beakers. These transducers were connected to a sine wave generator and a variable wattage amplifier, and set into oscillation at the appropriate frequency. For the early trials, too high a power was used, resulting in the loss of a number of transducers.

For budgetary reasons, therefore, the power was limited to about 25 W for these trials. The majority of the pulp used in this study was prepared at the James River Corp. Green Bay Mill secondary fiber laboratory from 100% laser printer furnish. For comparison, some laser-free pulp was also obtained from the mill's deinking process, prior to the addition of any chemicals.

The laser pulp was poured into a 1230-mL stainless steel beaker measuring 5.7 in. (14.5 cm) in height by 4.1 in. (10.5 cm) in diameter. It was subjected to various amounts of ultrasound, and then washed by 0.180-mm screens and a laboratory flotation washer provided by Doshi and Associates, Inc.

To wash the pulp, the slurry was first transferred to a 4-L beaker. Cold tap water was added to the slurry, increasing its volume to 4.0 L. A magnetic stirring bar was added, and the slurry was mixed for 5 min. The slurry was then poured onto the screen and shaken by hand until no more liquid could be drained from the screen. This procedure was repeated two more times, and the remaining pulp then transferred to the 900-mL flotation cell. This procedure was designed to simulate a commercial deink washing system, such as using vertical screens.

After the pulp was placed in the flotation cell, the air supply was turned on, and the cell was filled with cold tap water until it just overflowed the lip of the cell. The air flow was adjusted, if necessary, until the pulp exhibited a rolling motion within the cell. 250-mL of additional cold tap water were passed through the pulp over a 10–15-min time period by dripping the water into the cell from two burettes mounted above; the excess water was allowed to overflow and was collected for disposal. In this manner, a commercial dispersed air flotation system was simulated without using, however, any chemicals to promote foaming or flocculation.

The pulp remaining in the cell was then transferred by vacuum into a 1-L filtering flask and stored in a nalgene

bottle until evaluation. No attempt was made to optimize either the washing or the flotation procedure in any of these tests.

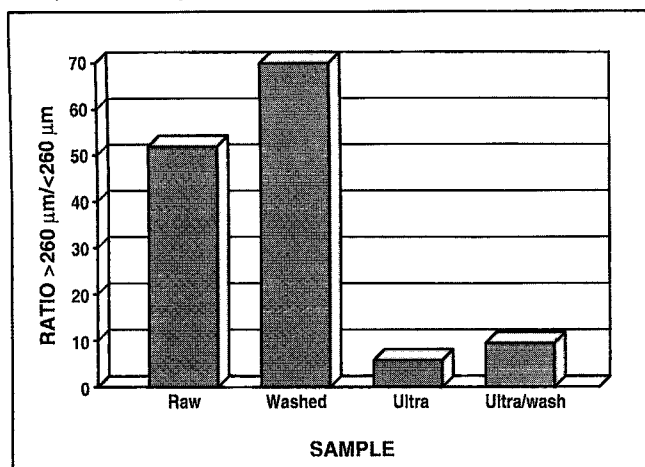
The success of the deinking was determined at the James River laboratory. In preliminary studies, the ultrasound was used to deink laser-free pulp and the results were evaluated by measuring the brightness using a Macbeth 1500/PLUS Color Measurement System. Brightness is a measurement of the ratio of the intensity of the reflected-to-incident white light for handsheets made of the various pulp samples. Unfortunately, brightness was not a suitable evaluation test for the laser-printed pulps. The very small ink particles gave a grayish color to the laser-free pulp. Due to the much larger sized ink particles, the laser-printed handsheets always appeared, in contrast, quite bright; in fact, even without any treatment, they exceeded the goal of the existing deinking process. It was therefore necessary, instead, to evaluate the sheets by determining the dirt count.

The dirt count was calculated by an Omnicon 3600 Image Analysis System. The handsheets were placed on a table beneath a camera and bathed in high-intensity white light. The paper reflects the majority of the light, but the black ink particles absorb some of the light and appear on the imager as green spots. The camera takes a picture of the handsheet; this picture is then digitized and fed into the microcomputer.

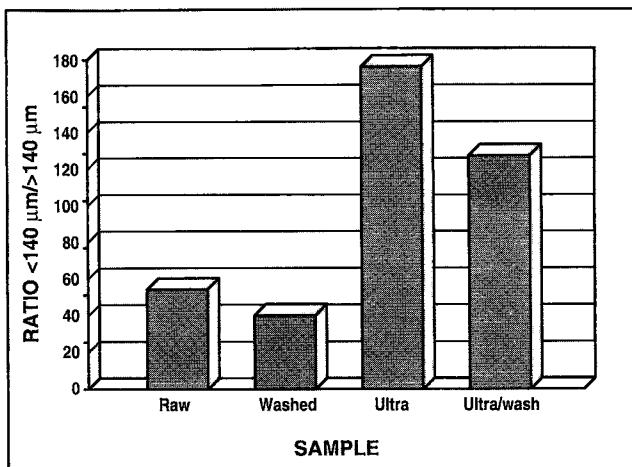
To find the percent area covered, the total area of the ink particles is calculated and then divided by the area scanned. The equivalent diameter information is also tabulated to give the distribution of particle sizes from 0 to 0.400 mm, the number of oversize (>0.400 mm) particles, and the maximum and minimum particle sizes. In every case, dirt counts were made of both sides of the handsheets and the reported results are the average of these two values.

Two different classes of ultrasound

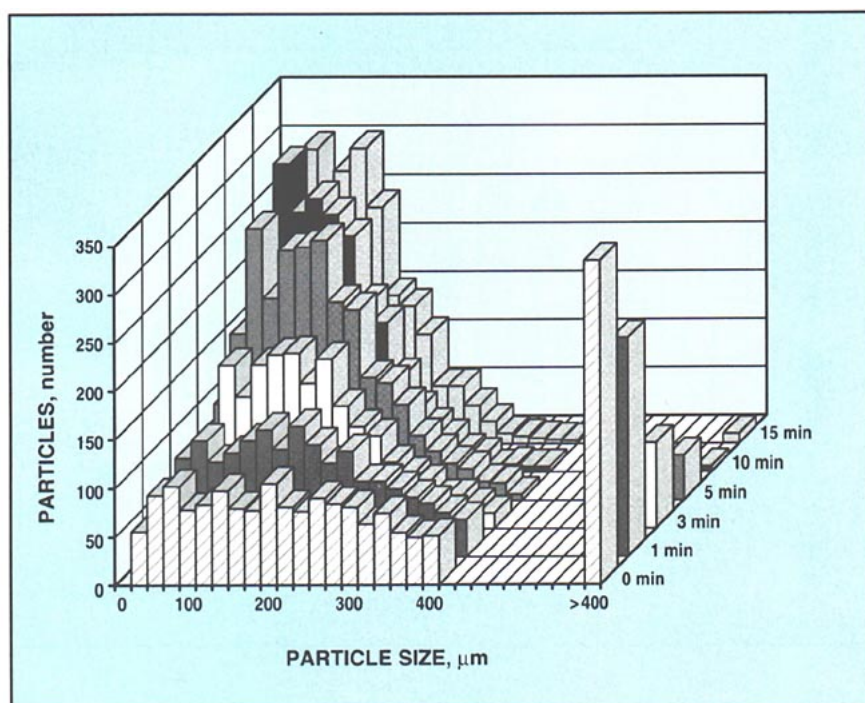
6. Separation study: effect of ultrasound on particle size



7. Separation study: floatable vs. nonfloatable particles



8. Particle distribution vs. process time



deinking tests were performed. The first type of test investigated the impact of various operating variables on the ability of the ultrasound to deink the fibers. Included in this category were the effect of pulp consistency and processing time. In all of these tests, the ultrasound treatment was followed by identical washing and flotation steps. The second class of tests analyzed the mechanism of the ultrasound deinking process. To evaluate the effect of the ultrasound on the

mechanical properties of the fibers, the Canadian Standard Freeness (CSF) was measured.

Deinking efficiency

Processing time

One liter of 1.0% consistency laser pulp was ultrasounded at 20 kHz by the Fischer Dismembrator for 0, 1.0, 3.0, 5.0, 10.0, and 15.0 min, then washed and floated. Two handsheets were pre-

pared from each pulp sample, so that we could investigate the reproducibility of the dirt count analysis.

To minimize any fluctuation between pulp samples, all of the data are compared to the results obtained for the washed/floated only pulp. **Figure 3** relates the increase in the percentage of dirt removed with the process time, where:

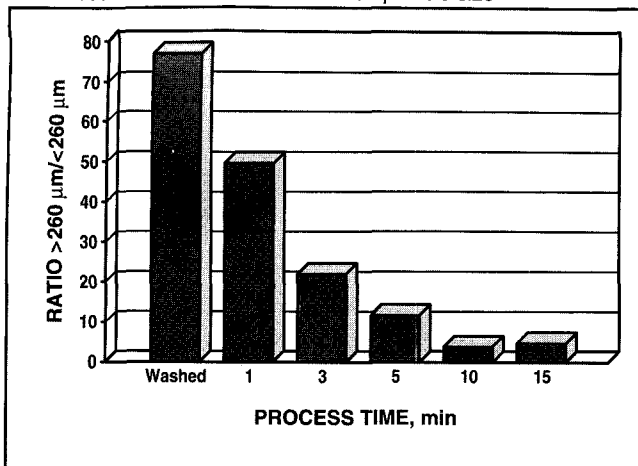
$$\% \text{ Dirt removed} = 100 \{ 1 - \text{Area covered by ink after ultrasound} / \text{Area covered by ink without ultrasound} \}$$

As can be seen, the portion of the handsheet area covered by ink particles decreases (the dirt removed increases) with processing time until the 10.0-min treatment time is reached, where it stabilizes at about 20% of the area covered by ink particles in the untreated pulp.

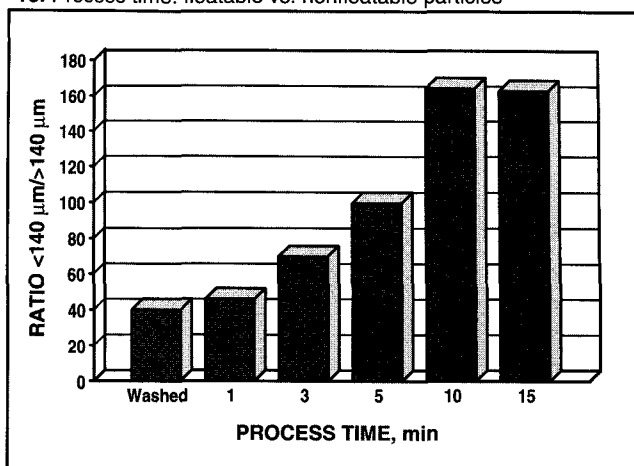
Consistency

The optimum consistency of the pulp determines where in the overall deinking process ultrasound treatment could occur. For a stand-alone process, a higher consistency process would undoubtedly be more economical. However, the ultrasound cannot cause cavitation in high-solids-content pulp slurries. The impact of varying the consistency between 0.50 and 2.0% is shown in **Fig. 4**. In all of these trials, 1.0 L of the pulp was ultrasounded at 20 kHz for 5.0 min using the Dismembrator.

9. Process time: effect of ultrasound on particle size



10. Process time: floatable vs. nonfloatable particles



As is indicated by the dirt removed, consistencies of 0.50% and 1.0% are more effective than the higher consistencies. Increasing the consistency to 1.5% significantly lowers the effectiveness of the ultrasound treatment, but increasing it further to 2.0% has little or no effect. The slightly lower dirt removal at 1.5% compared to 2% is not significant given the reproducibility of the various trials.

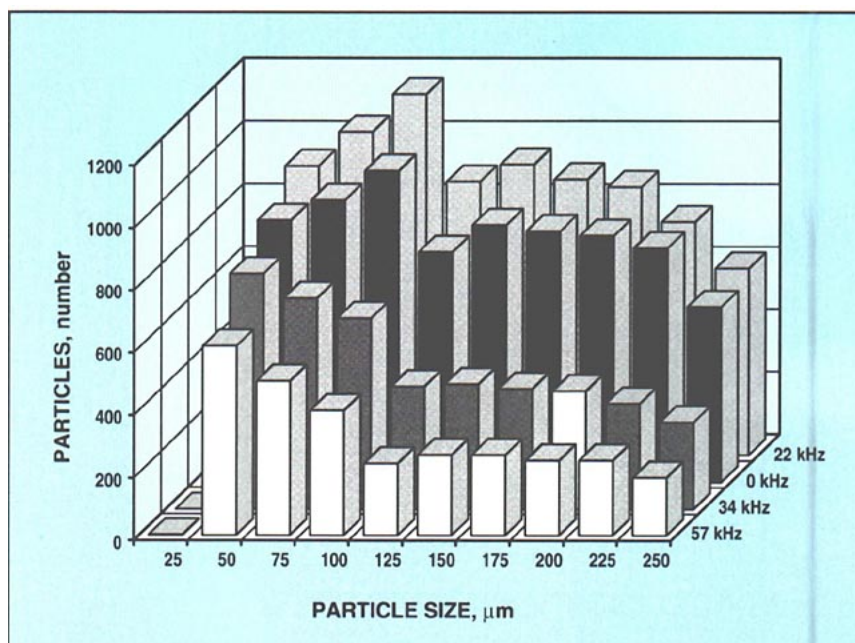
Deinking mechanisms

Separation of process steps

To isolate the effects of the different process stages, i.e., the ultrasounding and the washing/flotation, a series of tests were made on 1.0 L of pulp. These tests included no treatment, washed/floated only, ultrasounded only, and ultrasounded plus washed/floated. The ultrasound treatment consisted of 5 min of processing using the Dismembrator on a 1.0% consistency pulp. **Figure 5** illustrates the resulting particle distributions.

Figures 6 and 7 illustrate this effect of particle size from another perspective. **Figure 6** depicts the ratio of larger ($>0.260 \text{ mm}$) to smaller ($<0.260 \text{ mm}$) particles. It is apparent that the ultrasound treatment dramatically reduces the number of large particles and/or increases the number of small particles. The subsequent washing/flotation increases the ratio both for the

11. Particle size distribution—washed



untreated and the ultrasounded pulps since it removes some of the smaller particles.

Figure 7, on the other hand, considers the particles which can be optimally washed and floated, those $<0.140 \text{ mm}$. It should be no surprise that ultrasound treatment produces more small particles, and that washing and floating removes many of them. This figure clearly portrays how a larger percentage of small particles is removed from the ultrasound treated pulp, likely because the newly produced particle fragments are not

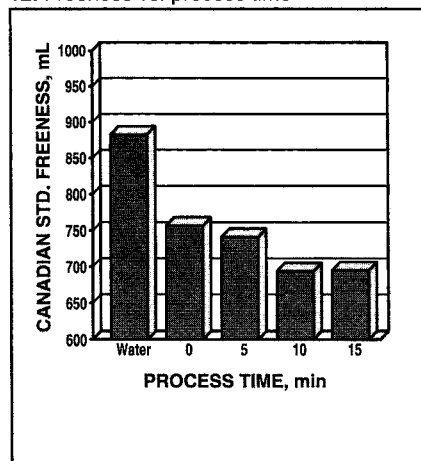
bound to the fibers.

Time-dependence of the particle distributions

The particle distributions that were determined for the 0-, 5-, 10-, and 15-min treatments in the above time study were then examined to see if they substantiate the above ultrasounding mechanism. **Figure 8** illustrates the relationship of the particle distributions with ultrasounding time.

As was portrayed in Figs. 6 and 7, **Figs. 9 and 10** depict the ratio of large-

12. Freeness vs. process time



to-small (>0.260 mm/ <0.260 mm) particles and the ratio of very small-to-larger (<0.140 mm/ >0.140 mm) particles, respectively. In Fig. 9, it is unmistakable how the percentage of large particles decreases with treatment time until it stabilizes after 10 min.

Similar results can be observed in Fig. 10, which illustrates the increase in the number of small particles. In both of these figures, the slight increase in dirt at 15 min compared to 10 min is within experimental reproducibility.

These results support the conclusions observed in the separation of process steps study; ultrasound treatment at 20 kHz very effectively splits apart particles 0.260 mm or greater in diameter. In fact, it seems that the larger the particle the more effective the ultrasound is in breaking it apart. These particles then fall into the size region where washing and flotation can effectively remove them. The inability to split apart more of the large particles after 10 min may be due to the scarcity of the large particles with respect to the cavitation bubbles.

Effect of other ultrasound frequencies

Using the bottom-mounted transducers, a series of tests was then made to determine the effect of ultrasound frequencies other than 22 kHz. Figure

11 summarizes the results. In all cases, similar pulp was irradiated for 120 min at a low power level (23 W), then washed and floated.

A comparison of the distributions of particles which have a diameter less than 0.250 mm illustrates significantly fewer smaller particles are present after treatment with either 34-kHz or 57-kHz ultrasound than are present after treatment with 22 kHz or with no treatment. This suggests that the higher frequencies break apart and loosen the smaller particles, which are then washed from the system. The increase in the number of particles in this size range after treatment with 22 kHz (compared to no treatment) is likely due to the splitting apart of the larger particles to form smaller particles, as was previously observed. Our less than optimum washing scheme leaves some of these newly formed small particles remaining in the pulp.

Further investigation of this aspect of the study is ongoing.

Fiber property modifications

To measure the effect, if any, of the ultrasound on fiber properties, particularly the possible fibrillation, the Canadian Standard Freeness (CSF) was measured. Figure 12 depicts the change in CSF as a function of processing time. Clearly, there is a significant increase in fibrillation (a decrease in freeness) due to the ultrasound treatment.

Deinking of laser-free paper

As was illustrated in Figs. 1 and 2, laser-free pulp contains significantly fewer and smaller ink particles than laser pulp. Ultrasound treatment has, nevertheless, virtually the same effect on the particles, as can be seen in Fig. 13 for 1% consistency pulp which has been ultrasounded for 5 min. As with the laser pulp, the main effect of the ultrasound is to split apart the larger particles.

Figure 14 contrasts the results of ultrasound treatment on laser and la-

ser-free pulp. For this example, 1 L of 1% consistency pulp was processed for 5 min at 20 kHz. Since particles smaller than about 0.08 mm are not observable to the naked eye, the ink particles which remain after ultrasound treatment of laser-free paper primarily give a slight color to the pulp; there are very few visible dirt specks. A measure of the brightness after ultrasound treatment verified that, for laser-free furnish, the pulp was comparable to pulp deinked by chemical methods.

Economic and process considerations

The increased efficiency with decreasing consistency, in addition to the current industry trend of not separating laser-print papers from ordinary office papers, suggests that the ultrasound process might be most effectively used as a polishing step, located perhaps among the series of flotation units. Typically, the deinked pulp is at its lowest consistency at this point. Large transducers could be mounted at several points on the sides of the pipes connecting these flotation washers. The pulp should first traverse the lowest frequency ultrasound and, in subsequent treatment sections, be subjected to ultrasound energy of increasing frequency. The major operating cost for ultrasound deinking is the cost of electrical power.

The cost of large, heavy duty transducers is surprisingly inexpensive, especially when one considers the cost of the small, laboratory transducers used in this study. For example, one of the vendors we contacted markets a 3-ft-long 20-kHz transducer with a tank for less than US\$ 2000.

Conclusions

Ultrasound energy clearly can split apart and loosen laser-print ink as well as conventional inks. The preliminary results indicate that using a spectrum of ultrasound frequencies will permit the removal of all sizes of ink particles.

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The process, therefore, appears hopeful as a stand-alone treatment for laser-print papers or, more likely, as a polishing step for mixed office paper grades. **[1]**

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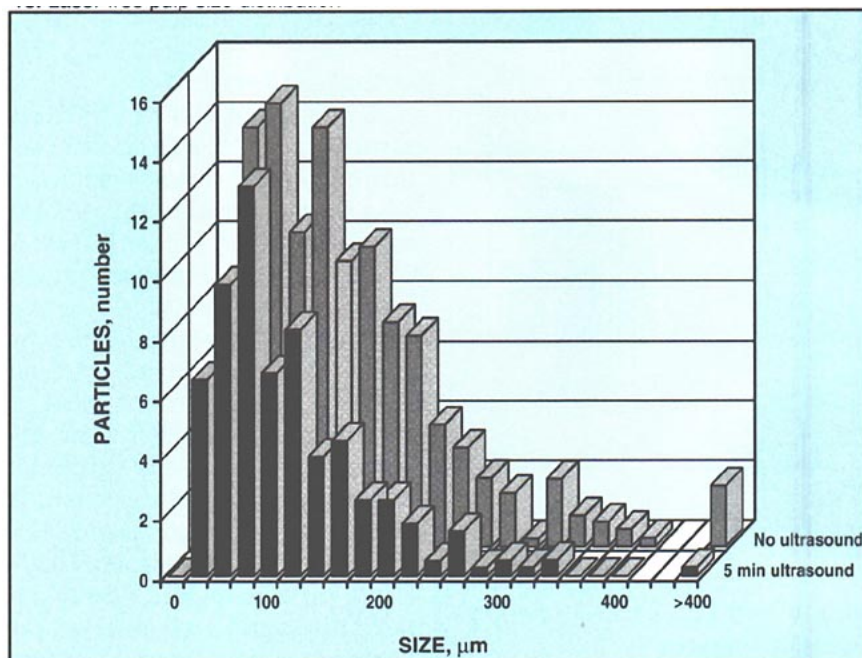
We sincerely thank our colleagues at the James River Corp. Green Bay mill for permitting us to use their facilities and especially Dan Sumnicht and Angie Lewandowski for their assistance with the laboratory tests. This project would not have been possible without their cooperation and kindness. Special thanks also to Mahendra Doshi, Doshi and Associates, Inc., for the use of his flotation washer.

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13. Laser-free pulp size distribution



14. Laser vs. laser-free pulp size distribution (5-min ultrasound)

