Using ultrasound to deink xerographic waste

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ABSTRACT: Mixed office waste containing xerographic ink is becoming an important source of recycled fiber. This research examined the use of ultrasound to break down xerographic ink particles prior to removal by conventional flotation. The raw material was standard xerographic sheets printed with common toners (styrene–butadiene and styrene–acrylate). Preliminary tests were carried out with a 500-W piezoelectric transducer capable of batch processing. Results from these tests provided a base for subsequent tests with an ultrasonic whistle suitable for continuous, flow-through operation. Both systems reduced xerographic particles to sizes suitable for removal by flotation. Performance of the continuous system was strongly affected by treatment pH, the number of treatment cycles, and pulp consistency. Response-surface methodology was employed to determine the best operating conditions for the ultrasonic whistle. Results for the continuous, flow-through ultrasound system suggest that scale-up experiments should be pursued.

KEYWORDS: Comminution, deinking, flotation, mixed waste papers, operating conditions, parameters, pH, processing, reclaimed fibers, toners, ultrasonic frequencies, variables, xerocopy papers.

The paper industry is working hard to increase its use of post-consumer secondary fiber, with the goal of achieving a 50% recovery rate by the year 2000. To do this, it will be necessary to utilize mixed office waste at a higher rate than is currently practiced.

Recycled office waste has been used extensively in the production of paperboard grades, where lower-quality fibers can be tolerated *(1)*. To date, only limited amounts of these fibers have been used in the production of fine papers (1–2% of fiber furnish). The presence of xerographic printed material in office waste has been the main obstacle preventing wider use of these fibers. Xerographic printed material is difficult to deink *(2)*, resulting in unacceptable levels of residual ink in deinked fibers. If xerographically printed office waste could be successfully deinked on a large scale, it could become a very cost-effective component of many paper grade furnishes.

Problems with deinking xerography papers

Xerographic toners can be removed from fiber surfaces by repulping under alkaline conditions *(3)*. The difficulty arises in attempting to separate the dispersed ink particles from the fiber slurry. The size of the toner particles ranges from 40 μm to 400 μm, while flotation cells are only effective on particles ranging from 30 μm to 150 μm. This explains why flotation cells have achieved only limited success in toner removal. Washing methods that flush out very small ink particles (< 20 μm) have also been ineffective.

Centrifugal cleaning is another method of separation that sorts particles according to their specific gravity. Forward cleaners remove debris with densities greater than water. Even though the toner specks are denser than water, their shapes make them behave differently than

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most dense particles *(4)*. Consequently, forward cleaning has achieved only limited removal efficiencies for toner particles.

Finally, screening has also proved to be an inefficient method of separating toner specks from a fiber slurry. Toner particles are platelike in shape. While the diameters of these particles may be larger than the slots in a screen, their thickness is considerably less. If these platelike particles are aligned sideways, they can pass easily even through fine slotted screens.

It is clear that the physical characteristics of xerographic toner particles must be altered before conventional mechanical means of particle removal—screening, cleaning, flotation, and washing—can produce satisfactory removal efficiencies.

Current xerographic deinking systems

There are two basic methods for high-efficiency treatment of office-waste inks. Both methods alter toner particles for subsequent removal using one or more of the conventional mechanical techniques.

The first method involves the mechanical fracture of toner particles. This method breaks toner particles by mechanically rubbing or kneading them, similar to refining action. Efficient toner dispersion is best achieved with a high-consistency slurry (30–35%). The kneader used in this system is commonly referred to as a disperser. A disperser breaks down toner particles from the 150–300-µm range to particles of about 40 µm in size.

The second approach uses chemicals that agglomerate particles to above 400 µm in size. This changes their shape and density to the point where they can be efficiently removed by pressure screens and forward cleaners.

These two examples indicate that industry research has paid off to the extent that systems now exist to handle these difficult inks. However, the search continues for better methods of dispersing toner inks.

I. Process variables and limits for Phase I runs (batch system)

Process variable	*High*	*Low*
pH		
Disintegration	10.5	4
Ultrasonic	10.5	4
Intensity, W	300	100
Number of cycles	5	1
Consistency, %	2	0.5
Orifice		
Diameter, mm	2	3.5
Distance to horn, mm	2	8

II. Statistically significant process variables and optimum levels as determined by Phase I runs (styrene–butadiene toner)

Process variable	*Optimum level*
Number of cycles	5
pH	4
Intensity, W	300
Consistency, %	0.5
Orifice diameter, mm	2

Ultrasonic dispersion of xerographic inks

In a recent study, Norman and Sell *(4)* used ultrasonic energy to break up xerographic ink particles. Using a piezoelectric-crystal ultrasound generator, the researchers investigated the effects of ultrasonic frequency, pulp consistency, and processing time on ink-particle-size distribution. They found that the ultrasound treatment reduced ink particles to a size range where they could be removed from the slurry by flotation. However, this batch treatment is not feasible for use on a commercial scale.

The research reported here extends the findings of Norman and Sell *(4)* by investigating the feasibility of using a continuous, flow-through ultrasound generator for dispersing xerographic toner ink particles.

Experimental program

Experimental objectives

The primary objective of this research was to evaluate the performance of a continuous, flow-through ultrasonic generator in breaking up toner particles for subsequent removal during flotation. A second objective was to determine the optimum operating conditions for the continuous system.

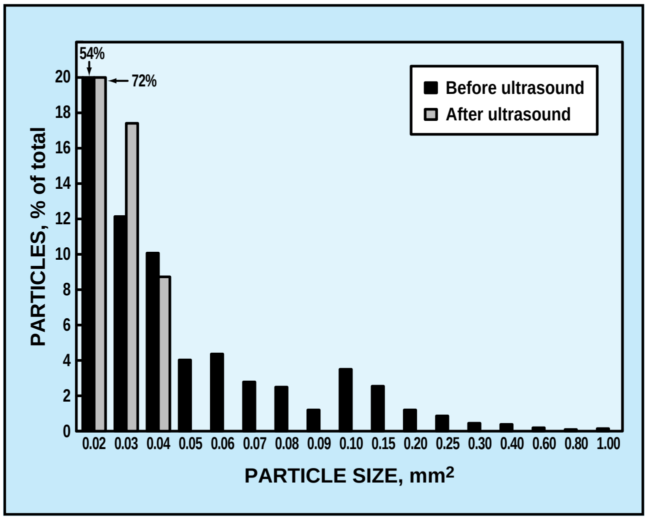
Experimental design

The experimental program was divided into three phases.

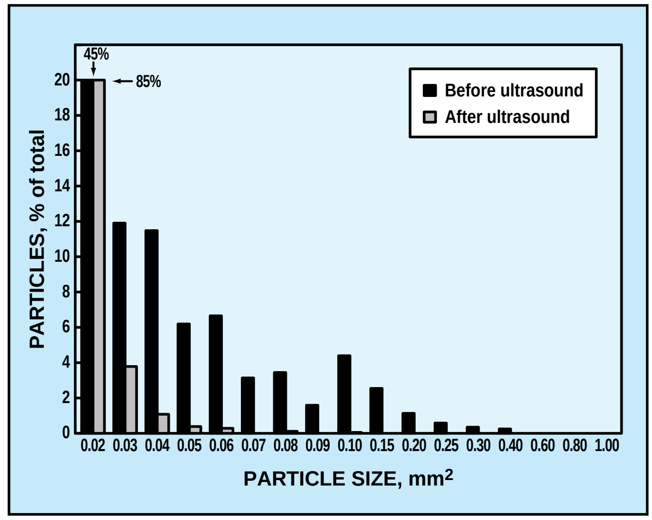
- Phase I—Construction and operation of a benchtop, ultrasonic, batch-treatment system based on a piezoelectric probe
- Phase II—Operation of a benchtop, ultrasonic, continuous-treatment system based on an ultrasonic whistle
- Phase III—Determination of optimum operating conditions for the benchtop, ultrasonic, continuous-treatment system.

Phase I. The experimental methods and procedures for the research project were developed during Phase I. The raw material was an office-waste slurry prepared from xerographically printed paper samples (Xerox Corp.). Papers printed with two common office-copier toners—styrene–acrylate toner and styrene–butadiene toner—were used in the

1. Size distribution of styrene–butadiene toner particles before and after ultrasonic treatment



2. Size distribution of styrene–acrylate toner particles before and after ultrasonic treatment



experiments. The printed black-area coverage was 10.9%.

Slurries were prepared by cutting xerographically printed papers into approximately 1.5-in. x 1.5-in. pieces and soaking them in water overnight. The papers were then defibered in a TAPPI disintegrator for 2 min. This process had practically no effect on particle-size reduction. Indeed, toner particles as large as a single letter could still be seen in the fiber slurry. However, the short defibering stage did eliminate excessive heat buildup and flow-rate variance resulting from orifice clogging during the batch-system ultrasonic runs of Phase I.

During Phase I, the office-waste slurry was funneled directly into a focused ultrasonic field, where the fiber and toner particles were bombarded with ultrasonic energy as they exited through a small orifice at the funnel. The ultrasound was generated by a 500-W piezoelectric transducer. This lab-type ultrasonic generator produced high-frequency (20-kHz) waves that were transferred to the slurry through a 0.5-in. horn.

The paper slurry was used in a series of experimental runs where seven process variables were set at the levels shown in **Table I**. In each run, handsheets were formed, pressed, and dried using procedures developed to retain dispersed ink particles. Twenty runs were done for each of the two toners investigated. Two handsheets from each experimental run were analyzed with a particle-size image-analysis system. The percentage of particles greater than 0.04 mm^2 and the average particle size were determined for each run.

A Plackett–Burman statistical experimental design *(5)* was used to determine the process variables with the greatest effects on the size of the treated toner particles. Followup experiments investigated the effects of pH and temperature on system performance.

Phase II. A continuous, benchtop, ultrasonic wastepaper treatment system was used during Phase II. Ultrasonic energy was generated using a whistle-type generator. The standard wastepaper slurries used in Phase I were also used in Phase II. Again, a Plackett–Burman statistical experimental design *(5)* was used to determine the process variables with the greatest effects on toner-particle disintegration. The variables with the greatest influence were carried on to Phase III.

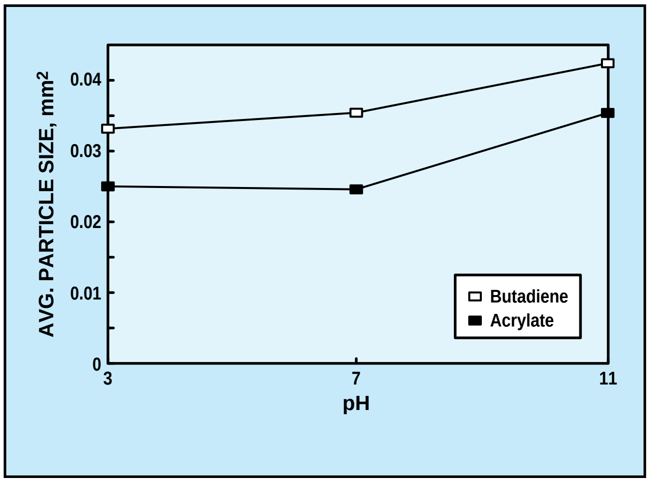
Phase III. Using the variables found in Phase II, additional experiments were carried out to discover the process conditions that would produce an ink-particle-size distribution suitable for flotation deinking. Box–Behnken factorial experimental designs *(6)* were used during Phase III. Mathematical models of the system were used to develop contour plots relating the size of the dispersed ink particles to process conditions. This approach is commonly referred to as a “response surface” method.

Results and discussion

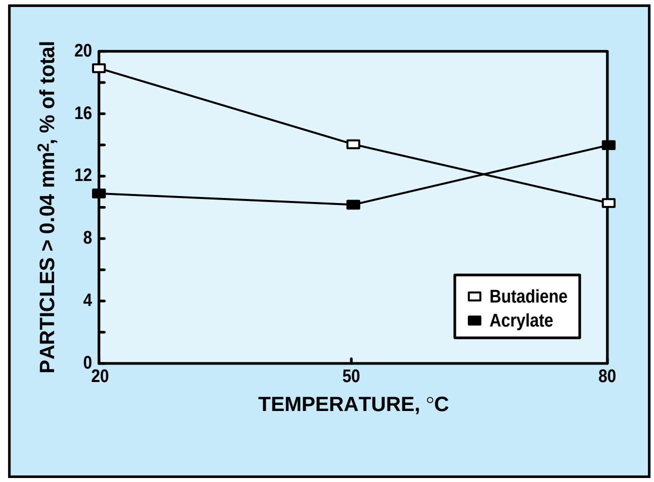
Phase I

The results of Phase I revealed that ultrasound intensity, the number of cycles through the system, pH, slurry consistency, and orifice size had statistically significant effects on both the average particle size and the percentage of particles greater than 0.04 mm^2. The number of cycles through the system had two-and-one-half times the effect of pH and ultrasound intensity, and it had four times the effect of slurry consistency. As the number of cycles through the system and ultrasound intensity increased, the particle size decreased. Consistency and orifice size had the opposite effect. The two different toners produced similar results. **Table**

3. Effect of pH on ultrasonic dispersion of styrene–butadiene and styrene–acrylate toner particles



4. Effect of temperature on ultrasonic dispersion of styrene–butadiene and styrene–acrylate toner particles



II contains the "best" or optimized process conditions derived from the first set of Phase I runs for styrene–butadiene toner.

Experimental runs were next carried out on the styrene–butadiene toner paper with the process variables set at the levels in Table II. Handsheets from these trials were compared with handsheets produced from fibers that had undergone only the initial defibering step but had received no ultrasound treatment. **Figure 1** displays the results for the styrene–butadiene toner. Prior to ultrasonic treatment, approximately 25% of the toner particles were greater than 0.04 mm^2. After ultrasound treatment, 100% of the particles were reduced to a size equal to or less than 0.04 mm^2. Moreover, before ultrasound treatment, 54% of the particles were less than 0.02 mm^2. After ultrasound treatment, 72% of the particles were less than 0.02 mm^2. (Note the greater width of the size classifications for the larger particle sizes along the *x*-axis. This was done to keep the *x*-axis in Fig. 1 to a reasonable length.)

Trials were also run for styrene–acrylate toners with the process variables set at their optimized levels. **Figure 2** illustrates the particle-size distributions before and after ultrasound treatment. Prior to treatment, only 69% of the particles were equal to or less than 0.04 mm^2. However, after ultrasound treatment, 99% of the particles measured were equal to or less than 0.04 mm^2. Moreover, prior to ultrasound treatment, only 45% of the particles were less than 0.02 mm^2. After ultrasound treatment, 95% of the particles were less than 0.02 mm^2. (Again, note the greater width of the size classifications for the larger particle sizes along the *x*-axis in Fig. 2. This was done to keep the *x*-axis to a reasonable length.)

Based on the results in Figs. 1 and 2, it is clear that ultrasonic treatment by a piezoelectric ultrasound generator is capable of reducing toner ink particles to sizes suitable for flotation removal.

Effects of pH and temperature on the ultrasound dispersion of xerographic ink particles. Under the same conditions, the two toners exhibited different responses to changes in pH. Whether dispersed in the TAPPI disintegrator or by ultrasound, the styrene–acrylate toner always fractured into smaller particles than the styrene–butadiene toner. This effect was visible to the naked eye.

It appeared that pH played a role in the different behaviors of the two toners. Therefore, separate pH trials were run. The pH was set at 3, 7, and 11, while the other process variables were kept constant (intensity 200 W, two cycles, consistency 1%, and orifice size 3.5 mm).

Contrary to the original hypothesis, the two toners exhibited the same relative behavior at the three pH levels investigated. Namely, the styrene–acrylate toner consistently fractured into smaller particles than the styrene–butadiene toner. This is illustrated in **Fig. 3**.

Toner/fiber pH effect. The original runs indicated that pH had a significant impact on ink-particle dispersion, with dispersed particle size increasing with increasing pH, as seen in Fig. 3. However, a followup pH experiment indicated that this was not a toner effect but, rather, a fiber effect, since the results were the same for both toners. The following explanation is offered for this phenomenon.

Upon addition of alkali, pulp fibers swell, opening fiber pores and aiding in the deinking process by dispelling ink particles from the fiber surface. These particles then float to the surface of the slurry, where they aggregate because of their hydrophobic nature. Finally, when a handsheet is formed, more of the floating toner particles end up on the sheet surface, where the image analyzer perceives them as large particles.

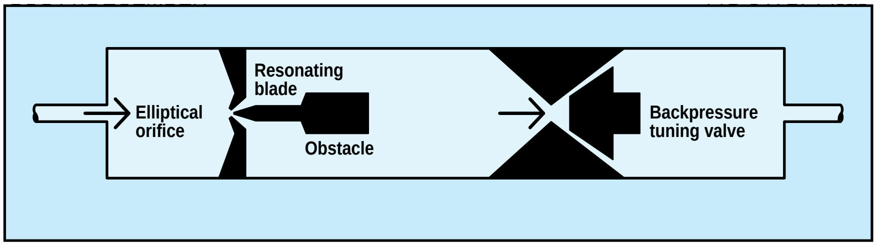
It seems likely that ultrasound would break up particles more effi-

III. Process variables and limits for Phase II runs (continuous system)

Process variable	*High*	*Low*
Number of cycles	8	2
pH	10	4
Intensity, psi*	300	150
Consistency, %	2.0	0.5
Orifice size, in.^{2}	0.007	0.003

*In the sonic whistle, the intensity is a function of pressure, and differences in backpressure levels are the controlling parameter for intensity levels.

5. Components of dispersing chamber with whistle-type ultrasound generator



IV. Process variables and limits for Phase III runs (continuous system)

Process variable	*High*	*Mid*	*Low*
pH	10	7	4
Number of cycles	12	7	2
Consistency, %	3	1.75	0.5

ciently if they were still attached to fiber surfaces. Ink specks that remain attached to the fiber would then be subjected to ultrasound treatment, in contrast to the particles that float to the top and agglomerate.

Temperature studies. Single-factor experiments were carried out to study the effect of temperature on ultrasonic dispersion of toner particles. Trials were run with the other variables held constant (intensity 200 W, two cycles, pH 7, consistency 1, orifice size 3.5 mm). Temperatures during the runs were 20°C, 50°C, and 80°C. These temperatures spanned the softening temperatures and glass-transition temperatures (T_g) of the two toners. Styrene–acrylate and styrene–butadiene toners both have T_g values of approximately 55°C. However, their softening points are quite different. Styrene–acrylate toners begin to soften at about 80°C, whereas styrene–butadiene toners soften at 90–100°C. Thus, 80°C was chosen to study the effects of temperature above T_g temperatures and close to the softening point of styrene–acrylate toner.

Figure 4 shows that the size of the styrene–butadiene particles decreased with increasing temperature. This makes sense, since the styrene–butadiene toner could not soften at the high temperature of 80°C during this experiment. On the other hand, ultrasound cavitation increases with increasing temperature in water-based fluids. Therefore, the efficiency of ultrasound in breaking styrene–butadiene toner particles was enhanced at elevated temperatures. The net result was a decrease in particles size with increasing temperature.

In contrast, the styrene–acrylate particle size remained fairly constant at 20°C and 50°C and then increased at 80°C, as seen in Fig. 4. This can be explained by the fact that at temperatures greater than 50°C, the styrene–acrylate particles begin to soften and agglomerate to form larger particles. This increased agglomeration tendency overrides the increased cavitational efficiency, resulting in a net increase in particle size.

Conclusions for Phase I. Phase I confirmed that ultrasound energy could reduce the size of dispersed xerographic ink particles to levels that should be removable using conventional flotation technology. Temperature and pH were found to have significant effects on process efficiency, as did slurry consistency and the amount and intensity of ultrasound treatment.

The ultrasound generator employed in the Phase I experiments would not be economically viable in a commercial operation because of its very high power requirements. However, the results of the Phase I experiments encouraged further experimentation with an ultrasound generator that required less power and had scale-up potential. This work was undertaken in Phases II and III.

Phase II

The Phase II experiments utilized a liquid-whistle ultrasound generator. **Figure 5** is a diagram of the whistle's ultrasound-generating chamber.

An office-waste slurry was fed through a funnel to a progressive-cavity pump that transferred the slurry to a dispersing chamber. The pump was driven by a 2-hp variable-speed motor with the capacity to pump 1–4 gal/min. Once the slurry was pumped through the dispersing chamber, it was recycled back to the funnel where it could be taken through another cycle.

The objective of the Phase II experiments was to identify the variables with the greatest effect on the process. The identified variables were then carried forward to the Phase III process-optimization runs. **Table III** lists the five significant variables that were identified in Phase II. As in Phase I, a Plackett–Burman statistical experimental design *(5)* was used to identify the significant variables.

The vibrating blade in the ultrasonic whistle subjects fibers to mechanical shear. It was necessary to determine the effect of this mechanical shear on toner particles before carrying out the screening runs. This was done by pumping slurries containing each toner through the whistle system under various pressures with the blade backed off so that no ultrasound was generated. The results indicated that some particle-size reduction occurred during this process. However, the extent of the size reduction was nowhere near that obtained when the system was operated under ultrasound-producing conditions. Therefore, it was concluded that the ultrasound played a major role in particle dispersion, and the Phase II runs were undertaken.

The statistically significant process variables were identified as pH, consistency, and number of cycles. The effects of intensity and orifice size were not statistically significant. These results compared well with visual observations made during the runs. For example, when the pH was increased, the fiber appeared to be more hydrated, and more ink was dispelled from the fiber to the slurry surface. The dispelled ink particles agglomerated and formed larger particles at the slurry surface. This was the same phenomenon observed during Phase I.

Finally, it was visually evident that as the number of treatment cycles increased, the particle size decreased. This effect was apparent up to about eight cycles, after which no more reduction in particle size could be observed visually.

Phase III

The Phase III experiments were designed to optimize the significant process variables identified during Phase II. **Table IV** lists the process variables selected for the runs and their levels. The ultrasound intensity was maintained at 250–280 psi and the orifice size at 0.007 $in.^2$ throughout the Phase III trials. Average particle size and percent particles >0.04 mm^2 were the response variables in the experiments. Both toners were investigated.

The optimum conditions for particle-size reduction were identified using a Box–Behnken *(6)* response-surface statistical experimental design. Full quadratic regression models were derived from the experimental results. These models were used to develop contour plots relating the size of dispersed ink particles to process conditions. The models were searched for maximum and minimum response points, and the optimum operating conditions were determined.

Only a summary of the Phase III results is presented here. A complete discussion of the response-surface data analysis is available in the literature *(7)*.

Styrene–butadiene toner. Analysis of the response-surface models revealed that the optimum operating conditions for the styrene–butadiene toner were pH 4.9, nine cycles, consistency 2.5%. Under these conditions, the average particle size was less than 0.02 mm^2, with 99% of the particles less than 0.04 mm^2. The system response was the most sensitive to pH and least sensitive to consistency.

Styrene–acrylate toner. Analysis of the response-surface models for styrene–acrylate toner indicated that the optimum operating conditions were pH 6.2, nine cycles, consistency 0.6%. As with the styrene–butadiene toner, under these conditions the average particle size was less than 0.02 mm^2, and 99% of the particles were less than 0.04 mm^2. Again, the system response was the most sensitive to pH and least sensitive to consistency.

Comparing both models, it appeared that a mixed toner furnish could be best treated at pH 5–6 for nine cycles at a consistency of 2.5%.

Summary and conclusions

The results indicate that pH and the amount of treatment (number of treatment cycles) are the critical operating parameters in the continuous, flow-through ultrasound system. The best ultrasonic reduction of toner particle size occurred at pH 4–6, in contrast to the alkaline pH normally employed in conventional deinking practice. As expected, particle-size reduction increased with increased amount of treatment. Slurry consistency did not exhibit a large effect over the range examined in this study.

It was concluded that ultrasound—generated by either a piezoelectric transducer or a continuous, flow-through ultrasonic whistle—can effectively reduce xerographic toner particles to a size range where they could be removed by flotation. This research suggests that further scale-up work should be pursued with the continuous, flow-through ultrasound system as a step in the deinking of mixed office wastes. The batch piezoelectric ultrasound system, while providing valuable experimental information, would be too costly to operate in a production situation. TJ

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