PROJECT SUMMARY
IMPACT OF IRRADIATION ON THE RECYCLABILITY OF MAIL

Presented 16 May 2002 10:00 am
White House Conference Center

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Background

In response to the anthrax contamination that occurred in several East Coast locations in the autumn of 2001, the United States Postal Service (USPS) began using electron beam technology to eliminate possible contamination in U.S. mail. Initially, all mail passing through anthrax-contaminated USPS mail processing facilities was sent to one of two private facilities that could expose the potentially contaminated mail to this technology before it was delivered to USPS customers.

At present, mail delivered to the general public is no longer being irradiated. However, mail that is destined for most government offices in the Washington D.C. area continues to be diverted to the irradiation facilities for processing before being delivered.

The USPS has received a number of complaints and comments regarding the condition of the mail as it is delivered to these governmental offices. Specifically, there were concerns regarding changes in paper color and texture, consistency of the ink, and enhanced brittleness as an apparent result of the irradiation process. As the U.S. government is strongly supportive of office paper recycling programs, these changes brought concern about the impact that the irradiation process might have on these office paper recycling programs and the overall recyclability of paper after irradiation.

Purpose and Project Objectives

In early March 2002, the USPS assigned to Roy F. Weston, Inc. (WESTON®), as part of the National Environmental Services Contract between the USPS Environmental Management Policy and WESTON, to study the impacts of irradiation on the recyclability of mail. To provide laboratory services for the project, WESTON hired Integrated Paper Services, Inc. (IPS), an independent paper sciences laboratory based in Appleton, Wisconsin.

Printed on recycled paper

The purpose and project objectives of the project were to do the following:

- Analyze and document the physical and chemical changes that occur to paper during the electron beam irradiation process;
- Identify changes that would affect the recyclability of paper.

Project Scope

After the project was assigned, the scope of work was further refined on 15 March 2002 at a meeting attended by representatives of the USPS, the White House Task Force on Recycling, the General Services Administration, IPS, and WESTON. The group's primary goal was to define the project and identify the sampling and testing protocols to be used in investigating the changes that occur in paper during the electron beam irradiation process. Tests to be performed on irradiated and control papers included gross appearance and handling, instrumental tests of paper strength and brightness, molecular chain length of cellulose, and microscopy. In addition, the papers were subjected a laboratory simulation of recycling, remade into paper, and tested again.

Sample Selection

Three samples were collected for the irradiation study. The majority of paper found in circulation in the USPS system is produced from chemical pulps. To simulate this fraction of the mail, a common copier grade paper was selected (Sample 1). This was a 77 gram/meter^2 grade manufactured under the Hewlett Packard name made up of 54% hardwood bleached kraft pulp and 46% softwood bleached kraft pulp.

Because the paper, which circulates generally, contains printing, 75% of the sample was printed using both laser and copier toners. The printing pattern contained both lettering and block designs. A quantity of each type was folded in thirds to simulate folded letters. The paper was equally divided, sealed into bags for shipment to the irradiation facility. Half of the paper was irradiated with the same level as used for the mail delivered in the Washington D.C. area. The other half was not irradiated and was used as a control sample.

Another common material found in the mail is magazine/flyer paper. These papers are chiefly made up of mechanical fibers and contain a high level of filler and or/coating. The main differences between mechanical fiber and chemical fibers are the lignin content and mechanical deformation. Mechanical fibers are typically crushed during processing and contain all of the lignin originally present in the wood. A 53-gram/meter^2 supercalender A grade (SCA) produced by Stora Enso North America was selected (Sample 2). Seventy-five percent of the SCA grade was printed via the rotogravure process. The sample was shipped directly from Stora Enso to WESTON. Half of the sample was packaged as a control sample by WESTON and half of the sample was packaged to be irradiated.

WESTON collected a mixed mail sample to simulate the mixture circulating throughout the mail system (Sample 3). This sample contained several envelopes, newsprint, copy paper, magazines, etc. Each component in the mixed sample was cut in half to make up the control and irradiated samples.

Exposure

WESTON hand delivered the samples (both the samples to be irradiated and the control samples) to the Ion Beam Applications (IBA) facility in Bridgeport, New Jersey, on 4 April 2002. The samples for irradiation were unpacked and packaged to mimic methods the USPS uses, while the control samples remained in their original packaging. This included placing the paper samples in USPS tubs, inserting the tubs inside corrugated cardboard sleeves, wrapping the sleeves and tubs inside a black plastic bag, sealing the bags inside corrugated cardboard boxes, and placing the boxes on stainless steel trays on the process conveyor for exposure to the electron beam. The control samples remained outside the process area and were not exposed to the irradiation process.

After an initial ½-hour in the system, the boxes emerged from the conveyor, were turned over (top to bottom) and sent through the process for another half hour. Information concerning the exact exposure time and level of exposure is not available due to security concerns.

Following the irradiation process, the exposed sample packages were unpacked and the following conditions were noted by Tom Hanzely of WESTON:

- All samples were hot (estimated at 160^{o} F).
- There was a discernible “hydrogen peroxide” odor.
- Significant amounts of moisture, which condensed on the inside of the plastic sample bags, were produced.

The samples were then rebagged, sealed to match the control sample packaging, and repackaged for shipment to IPS for analysis.

Gross Appearance and Handling

The samples arrived at the IPS laboratory on 5 April 2002 and were unpacked for an immediate visual inspection and comparison of the irradiated samples to the control samples.

Observations made by IPS staff indicated that the irradiation process visually degraded the samples. Each sample was very brittle when folded and would typically break when folded only twice. The printed samples of the copy paper would tear when attempting to unfold the paper due to adhesion between paper plies and the melted toner ink. The samples were also visibly yellowed, indicating a loss in brightness.

Paper Strength Properties

Several tests were made on paper strength and showed a significant decrease in the irradiated samples. Tensile (the resistance of paper to fracture when pulled from each end), tear (the resistance of paper to propagate a tear), and bursting strength (resistance of paper to penetration) are very common methods used in the industry to measure the overall strength of paper. Tensile, tear, and burst strengths for both samples 1 and 2 were lowered approximately 30%, 50%, and 40%, respectively. The results of these tests indicate a significant strength loss due to irradiation. The loss is especially key in the copier paper, which would typically be purchased as recycled fiber to provide strength in the papermaking process.

Another property that is very important for several types of paper is flexibility. This is measured by the folding endurance test and paper stretch during the tensile test. Folding endurance, which measures the number of folds the paper can withstand before breaking under a constant weight, was lowered over 90% in both samples 1 and 2 compared to the control samples. Paper stretch was lowered approximately 50% in both samples as well. The decrease of both properties in the CD (cross-machine) direction in the mechanical sample is low due to the lower overall level of these properties in the nonirradiated material. The results of these tests indicate the brittleness that exists in the paper after irradiation.

Note that the heterogeneous nature of sample 3 prevented the direct measurement of its physical and strength properties.

Optical Properties

Paper brightness and color are vital properties when establishing a price at which a particular grade of paper will sell. Brightness (reflectance at 457 nanometers) measures the overall whiteness of a sheet of paper. The copy paper brightness was lowered over 10%, and the SCA paper brightness was lowered almost 5% due to irradiation compared to the control samples. The brightness drop is crucial for recycling because lower-brightness material will require more bleaching chemicals in the pulp mill. Analyzing the paper for color showed that each sample had an increase in the yellow and green level in the paper. Paper typically yellows as it ages, and it appears the irradiation significantly accelerates the process.

Note that the heterogeneous nature of sample 3 prevented the direct measurement of its physical and optical properties

Laboratory Simulation of Paper Recycling

A portion of each of the three paper samples from both the irradiated and control groups was repulped (dispersed in water) to simulate the initial processing that takes place in a recycling facility. Once repulped, the samples were run on a PFI mill. The PFI mill simulates the refining process in a paper mill. Refining cuts and fibrillates pulp fibers to achieve proper drainage on the paper machine and the desired properties of the finished paper. Pulps are normally evaluated after five levels of PFI refining. Because of the low strength of the irradiated samples, only three-point PFI curves were developed since further refining would provide drainage levels unusable in the paper industry. The initial freeness levels for the irradiated samples were lower than the control for all three samples; specifically, the freeness was 7% lower for the copy/laser-printed paper and 18% lower for the SCA (mechanical) paper. Low freeness is indicative of an excess of fiber "fines" (small fiber particles); fines require a paper machine to be run more slowly. During refining, the freeness level also dropped significantly faster in the irradiated samples than the control samples.

Physical and Optical Testing of the Recycled Samples

Handsheets, small sheets of handmade paper, were prepared from each of the samples (both irradiated and control) following the simulated recycling process. These sheets were used for physical and optical testing. Approximately the same strength loss was obtained with the repulped and refined samples as in the original paper samples discussed earlier. The mixed mail sample (sample 3) appears to follow the same trends in its strength loss due to irradiation as the copy paper and SCA samples (samples 1 and 2) followed.

A brightness drop was measured in the handsheets of samples 1 and 2. The brightness testing of sample 3 handsheets revealed no difference in brightness due to irradiation. However, these results are skewed due to the high levels of contaminants (dirt, ink, etc.) in the handsheets.

Ink Removal During the Recycling Process

Flotation trials were conducted for all three samples to simulate the ink removal that occurs during the recycling process. The ink removal efficiency was very poor in the irradiated flotation trials for all three samples. To determine ink removal efficiency, dirt count was measured on handsheets made after flotation experiments with the control and irradiated pulps for each sample. The overall ink removal efficiency was 44% lower for the copy paper, 10% lower for the mechanical paper, and 97% lower for the mixed mail sample after irradiation compared to the control samples. The lower ink removal efficiency caused by irradiation would limit a mill's ability to achieve the brightness target of a particular grade if irradiated material was used in the process.

Chemical Properties

Chemical tests were performed to evaluate pH and pulp viscosity of each sample. Cold extraction pH was measured for all three samples (both irradiated and control) before and after irradiation. A significant drop in pH after irradiation was measured for each sample. The drop in pH indicates the formation of acids in the paper during the irradiation process. Acids form during the yellowing process as paper ages. Since the irradiation process yellows the paper, it is logical that acids are forming in the sheet as well. Irradiation may also be forming free radicals by reacting with the water present in the paper. Free radicals form acids when they react with the cellulose in the paper.

Pulp viscosity correlates with the degree of polymerization in the individual carbohydrate chains, which make up the cellulose in paper. A low viscosity suggests shorter carbohydrate chains and typically lower pulp strength. In samples 1 and 3, the viscosity level dropped over 84% after irradiation. The viscosity was 69% lower in sample 2 due to irradiation. These results indicate that chemical attacks, which are resulting in lower pulp strength, are occurring in the carbohydrate chains in the cellulose during irradiation.

Fiber Properties

Several methods were used to investigate changes in the physical nature of the fibers in each sample. Scanning electron microscopy (SEM) was used to take close-up photos of the paper surface to look for changes in both the fibers and the printing. No noticeable changes in the fibers were found in the photos of all three samples. However, the toner inks used to print the copy paper (sample 1) were quite different after irradiation. In the photos, it appears that the toner ink had melted and agglomerated into a smoother print matrix on the paper surface. The larger ink particles formed by melting the toner may impact their removal efficiency during flotation deinking. No changes in the rotogravure ink used to print sample 2 were found in the SEM photos.

A Kajaani FS-100 was used to determine the average fiber length in all three samples. The Kajaani FS-100 uses an optical sensor utilizing polarized light to measure the length of each fiber in a sample as it passes through a very narrow tube. In all three samples, the average length of the fibers in the irradiated sample decreased slightly compared to the average fiber length in the control sample. A small increase in the percentage of fines was also observed in the irradiated samples. In this technique, fines are defined as fibers shorter than 0.1 millimeter in length. The level of fines in a sample is significant because the fines not retained in papermaking lower the overall yield in the recycling process. The Kajaani data seem to indicate that the fibers weakened by irradiation break down into smaller fragments during repulping.

The level of fibrillation was studied under a light microscope for each sample. Fibrils are tiny, hair-like structures that protrude out of a fiber typically after refining. The fibrils are actually pieces of the fiber that are breaking away from the fiber surface. A slightly higher level of fibrillation was determined in the irradiated samples than in the control samples. The brittleness of the fibers may increase the fibrillation of the fibers during repulping and refining which increases sludge, decreasing yield.

Project Results

Analysis of the paper samples revealed significant deterioration of the physical, chemical, fiber, and optical properties due to irradiation. Strength properties in all three samples were decreased from 30 to 90% depending on the property compared to control samples. Ink removal efficiency by flotation was lower when the samples were irradiated. Viscosity and pH measurements indicate that significant chemical degradation of the cellulose had taken place in the paper fibers during the irradiation process. The brightness of the paper samples was lowered 5 to 10% due to yellowing. It appears that irradiation caused accelerated aging of the paper, resulting in yellowing and a decrease in strength properties.

The results of this study suggest that each of the samples analyzed can be recycled after irradiation. However, if the level of the irradiated paper makes up a significant portion of a mill’s raw material, perhaps 10% or more, the mill will have to compensate for the changes in the pulp properties according to IPS. Mills recycling irradiated mail will need to use less mechanical energy in refining to achieve the desired physical characteristics, but more bleaching chemicals will likely be necessary to provide the target brightness.

Most mixed office paper recycling collection programs will not be significantly affected by the presence of irradiated papers. Papers from typical mixed office paper recycling programs are generally directed toward paperboard and tissue mills rather than mills that demand the higher quality of white office paper recycling collection programs. These higher-end “white paper only” recycling programs as a rule do not generally include mail of any kind so as to avoid other contaminants, such as adhesives, groundwood content, and non-paper items, which are often present in mail.

Recommendations

Irradiated mail should be segregated from higher-grade recycled fiber if it is to be directed into paper mills producing grades of paper that demand high brightness and fiber strength. Use of irradiated fiber in significant quantities should be restricted to paper grades that do not demand strong, white fibers. These papers would include newsprint, paperboard, coated grades, and some tissues.

Office paper recycling programs that collect mixed office paper should not require any major changes to collection practices, but effort should be made to inform and work with local paper stock dealers to assure that irradiated papers will be beneficially utilized in the manufacture of recycled paper.

Study of the Impact of Irradiation on the Recyclability of Mail for the USPS

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