Surface sizing with cationic starch: its effect on paper quality and papermaking process

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ABSTRACT: The characteristics of starches used in surface sizing affect not only paper properties but also the efficiency of broke recycling processes. Oxidized starches commonly used for surface sizing tend to penetrate deeply into the paper structure, and this causes substantial reduction of opacity. Furthermore, oxidized starches act as anionic trash in the broke recycling process. To solve the problems associated with conventional size press starches, cationic starches were prepared and their effects on paper properties and papermaking processes were investigated. Results showed that cationic starches outperformed oxidized starches in opacity, brightness, print gloss, and ink density due to their better holdout property. Effectiveness in surface holdout of cationic starches was verified using a confocal laser scanning microscope (CLSM). Adsorption ratio of the cationic starch onto fiber was substantially larger than that of oxidized starch, and this decreased COD load and increased fines retention and strength. We observed improvements in stiffness, opacity, brightness, printing quality, and picking resistance when ammonium persulfate starch was substituted with cationic starch in the film transfer size press. Furthermore, diverse beneficial effects of improving white water quality, including the reduction of COD and cationic demand, were obtained. Surface sizing with cationic starch also gave better picking resistance and ink receptivity for coated paper than ammonium persulfate starch.

Application: To solve problems associated with oxidized starches, we prepared cationic starches and investigated their effects on paper properties and papermaking processes in laboratory and mill settings.

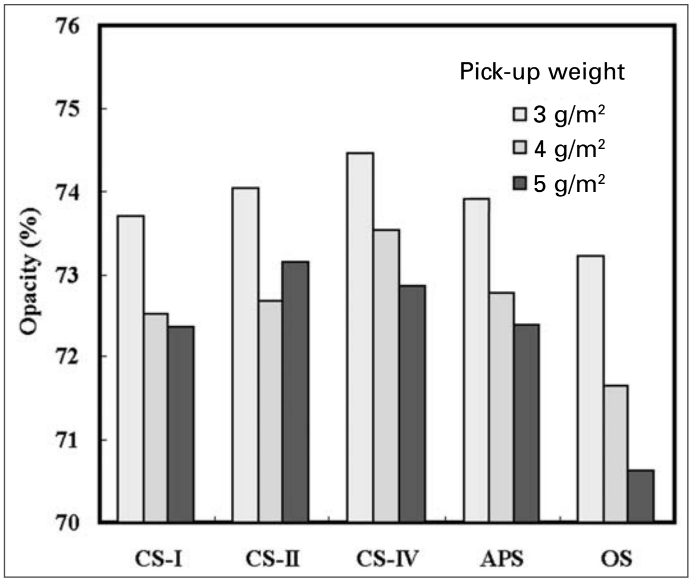
Traditionally, the paper industry consumes a large quantity of starches as internal and external additives. The purpose of external application of starch solution onto dried paper or surface sizing is to provide resistance to penetration of liquids, to give better surface properties, and to improve certain physical properties of paper sheet such as tensile strength and internal bond. To achieve this purpose effectively on today's high-speed paper machines, many technological developments have been made in surface sizing equipment during the last several decades. Klass [1] provides an excellent review on the development of size press technology. Among the new technologies, metering size presses provide solutions to some of the problems associated with the conventional size press, including improved surface starch application, increase of machine speed and runnability, etc.

Although surface sizing provides many advantages in improving paper properties without contaminating the wet end, a substantial proportion of the size press starches re-enter the papermaking system through the broke recycling process. It is quite common for more than 10% of the total production to become broke. If this contains 5% of surface sizing agents, more than 0.5% of the total furnish will consist of size press starch, which is introduced into the papermaking system along with the broke. Therefore, for a paper mill producing 1000 ton/day, 5 tons of size press starches are introduced into its papermaking system every day along with the dry broke. If the size press starch dissolves into white water while reslushing dry broke, severe contamination of process water and effluent will result, and this will limit the level of papermaking system closure.

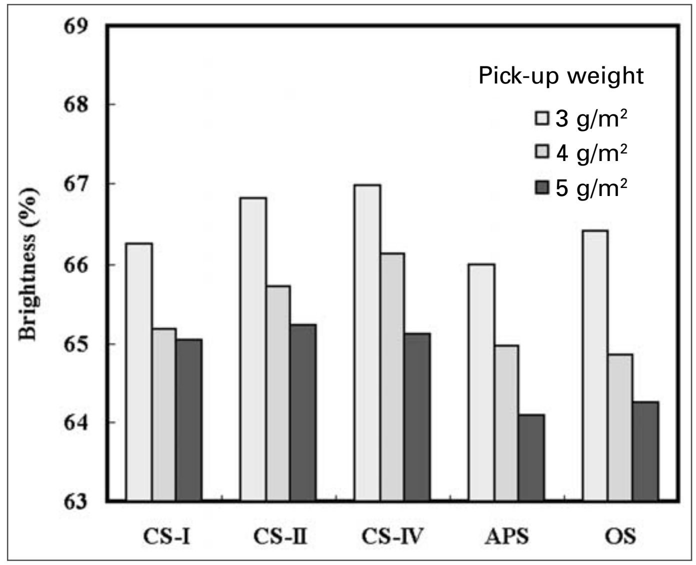
Starches for surface sizing range from the native in-mill converted products to preconverted starches. In-mill converted starches include enzyme-converted starch and ammonium persulfate (AP) converted starch. Oxidized starch, starch ethers and esters, and cationic starches (CS) are examples of preconverted ones, which have been characterized in other studies [2, 3]. If the level and type of size press starch used is inappropriate, it can result in loss of opacity and brightness, lower bulk, reduced machine speed, higher effluent losses, increased sludge build-up, and poor paper machine runnability [4].

During the past decade, many starch manufacturers have introduced cationic size press starches, mainly as an opportunity to reduce the starch load in white water and effluent [4-7]. This environmental benefit of cationic size press starches occurs because the starches remain tightly bound to the fiber even during the repulping of broke, thus preventing the starches being lost in effluent. Studies have shown that cationic starch use nearly halved the BOD level or effluent load [5, 7]. Improvement of paper properties, including ink-jet printability, sizing degree and sheet porosity, could be obtained by using cationic size press starch [6, 7].

The theory behind cationic size press starches is that they will bond with the anionic fiber in the manner of a wet end starch, which means they will form a stronger, more surface-oriented film. The effect is to raise the proportion of starch concentration at the surface to a level higher than is possible with conventional size press starches [4]. Kogler et al. used the concept of using interpolymeric interaction between cationic materials applied on the anionic charged paper surface to promote quick immobilization of coating colors [8]. Fast immobilization of cationic coating colors on anionic paper surface improves fiber coverage and printability. On the other hand, Remmer



1. Effects of starches on opacity. Base sheet opacity was 77.3%.



2. Effects of starches on brightness. Base sheet brightness was 69.6%.

and Eklund [9] showed that the degree of cationization of size press starches had no effect on absorption rate, which was unexpected since cationic starches were assumed to have a higher affinity for the paper fibers than the anionic starch and would therefore be expected to yield a lower absorption rate. In this study, however, Remmer and Eklund [9] defined the absorption rate as the total starch pickup that included starches that remained on the paper surface as well as those that penetrated into paper structure.

In most of the prior studies on cationic size press starches, simple comparisons were made on the effect of a cationic size press starch and other conventional starches on effluent contamination, paper properties, wet end chemistry, etc. [4-7]. In this study, we investigate the effect of four cationic size press starches with different degrees of substitution (DS) and viscosity on paper properties, wet end chemistry, and white water contamination, and compared the results with those of conventional ammonium persulfate and oxidized starches. Since a great amount of papers surface sized on the size press are used as coating base stock, we also examined the influence of size press starch types on the properties of coated papers. Finally, we evaluated the performance of a cationic size press starch on a high-speed paper machine equipped with a film transfer size press.

	CATIONIC STARCH				AMMONIUM PERSULFATE STARCH	OXIDIZED STARCH
	CS-I	CS-II	CS-III	CS-IV		
Brookfield viscosity (cPs)	11	26	11	21	9	9
DS	0.01	0.01	0.03	0.01	-	-

I. Characteristics of starches

EXPERIMENTAL

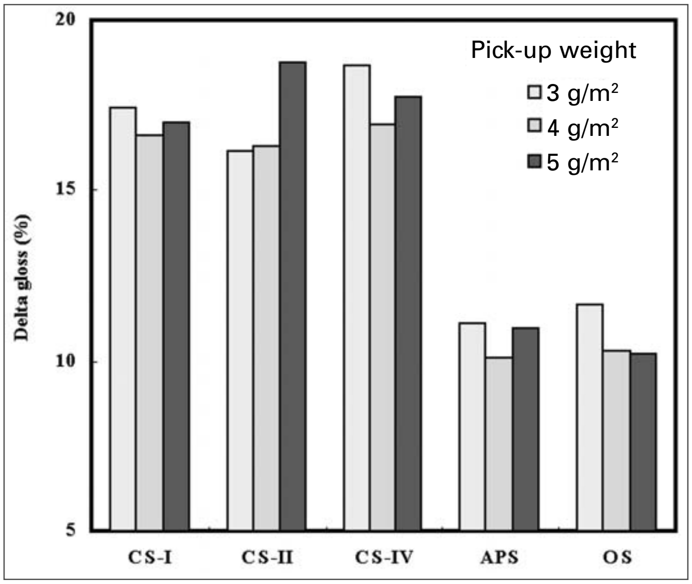
Starches

This study used four cationic starches with different DS and viscosity, ammonium persulfate starch, and oxidized starch, as shown in **Table I**. Ammonium persulfate starch was prepared by reacting 0.45% of ammonium persulfate with starch slurry at 10% consistency. Oxidized starch was prepared with the use of 3% sodium hypochlorite. Brookfield viscosity in Table 1 was measured using 10% starch paste at 50°C. As shown in Table 1, the Brookfield viscosity of ammonium persulfate and oxidized starches were 9 cPs. The viscosity of two cationic starches, CS-I and CS-III, were 11 cPs, while those of CS-II and CS-IV were 26 cPs and 21 cPs, respectively.

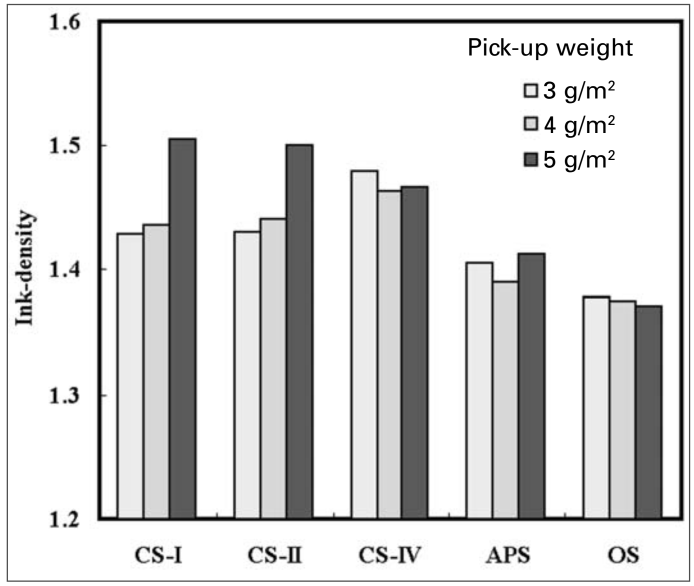
Surface sizing and property measurement

We prepared a starch slurry at 10% concentration and cooked it at a temperature of 95°C for 30 minutes. We kept the cooked starch paste in a constant temperature water bath at 65°C and used it for surface sizing. Coating base stock at a basis weight of 61 g/m² produced on a high speed gap former was used as substrate for the surface sizing experiment. The furnish of this base stock consisted of 80% hardwood bleached kraft pulp and 20% softwood kraft pulp. While producing the unsized base paper the starch solutions on the film transfer size press was substituted with water. The thickness of paper was 75 μm and contained 8% ash. Gurly permeability of the paper was 6.3 ± 0.3 sec. After surface sizing with starch solutions using an automatic rod coater, it was cylinder dried and lightly calendered on a soft calender. Starch pick up ranged from 3-5 g/m². Optical and strength properties of surface sized papers were determined according to TAPPI standards test methods. Delta gloss, trapping and ink density were measured after printing sample strips with cyan ink on RI printability tester. Picking resistance was determined after printing magenta ink with tack value of 18 on RI printability tester.

To investigate the penetration of starch solution into papers, we used confocal laser scanning microscope (CLSM) images for the cross section of the hand-



3. *Effects of starches on delta gloss. Delta gloss measured on base sheet was 7.2%.*



4. *Effects of starches on ink density. Ink density on base sheet was 1.29.*

sheets made from 70% bleached hardwood kraft pulp and 30% bleached softwood kraft pulp were obtained after surface sizing with starch solutions containing 0.1% of a fluorescence dye, Aridine-orange C.I. 46005.

Broke recycling

We examined the effect of starch types on broke recycling using surface sized handsheets. The starch dry pickup of these handsheets was 4±0.1 g/m^2. The disintegrated handsheets were mixed with stock consisting of 70% hardwood bleached kraft pulp and 30% softwood kraft pulp, both of which were beaten to 450 mL CSF. The ratio of the recycled surface-sized paper was 30%. We stirred the mixed stock for 60 min at 60°C in the laboratory pulper to extract starches.

Retention measurement and analysis of the filtrate

To evaluate the filler retention in the laboratory, we added 15%-25% of ground calcium carbonate (GCC) to the stock, and used 0.03% of a cationic PAM as a retention aid. The furnish pH was 7.5±0.2. We used a dynamic drainage jar (DDJ) to measure retention according to TAPPI Standard T261cm-90. Cationic demand and COD of DDJ filtrate were measured. Cationic demand was determined with PCD-03, and we used a COD reactor and DR-2000 spectrometer to measure COD. Adsorption of starch onto pulp fiber was evaluated by UV-Visible spectroscopy method. We made handsheets at a basis weight of 75 g/m^2 on a square handsheet former to examine the paper properties. Handsheets consisted of 30% surface-sized broke mixed with bleached kraft pulps, and 20% of GCC and 0.03% of PAM.

Coating

After dispersing No. 1 high brightness, ultrafine clay, 6 pph of starch and 10 pph of S/B latex were added as binders, and coating solids content was adjusted to 50%. We spread the coating onto plastic films to form, then determined the porosity by measuring the weight increase after saturating the coating with silicone oil. We determined brightness and opacity of the coatings by conventional methods.

RESULTS AND DISCUSSION

Effect of starch types on paper properties

To investigate the effect of the types of size press starches and the amount of their pickup on paper properties, opacity, brightness, gloss, and ink density were determined after surface sizing unsized base paper with cationic, oxidized and ammonium persulfate starches. The opacity of base sheet was 77.3%. As shown in **Fig. 1**, opacity decreased overall as starch pickup increased. Surface sizing with cationic starches caused less loss in opacity than surface sizing with oxidized starch. CS-IV, which was prepared by adding 3% of waxy maize starch, gave the least reduction in opacity. The high viscosity of CS-IV appeared to be a factor in that. However, CS-II, with a higher viscosity than CS-IV, causes greater reduction in opacity, suggesting that viscosity is not the sole factor for opacity reduction.

Surface sizing also decreased brightness, and brightness reduction increased as the amount of starch pickup increased (**Fig. 2**). The brightness of unsized sheet was 69.6%. Sheets surface sized with cationic starches showed relatively high brightness compared to the sheets surface sized with ammonium persulfate starch because ammonium persulfate starch is yellowish in color. Figures 1 and 2 reveal that the most significant loss of opacity and brightness occurred with oxidized starch. Surface sizing with cationic starches also gave greater delta gloss and ink density than other starches, as shown in **Fig. 3 and Fig. 4**.

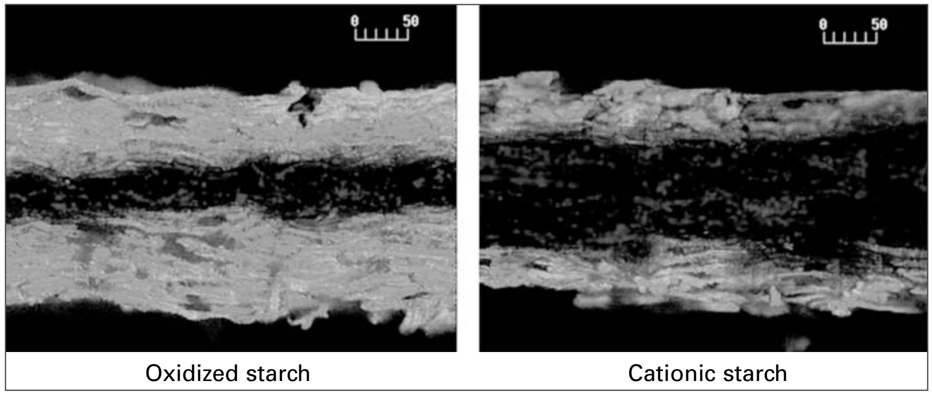
These results suggest that cationic starch penetrates less into paper after surface sizing. We used CLSM images of surface-sized handsheets to demonstrate the difference in penetration depth into paper for oxidized and cationic size press starches. CLSM images clearly showed that less penetration into paper occurred when cationic starch (CS-III) was used for surface sizing, i.e. more starch remained on the paper surface (**Fig. 5**). Cationic starches tend to stay on the paper surface rather than penetrating into the paper structure because of the electrostatic interaction between negatively charged fibers and positively charged starch molecules [4]. Oxidized starches tend to penetrate into paper. In other words, when cationic starch is used in surface sizing more internal pore structure of the paper can be preserved. This causes less reduction in opacity and brightness after surface sizing. It also improves printing gloss and ink density.

One of the main objectives of surface sizing with starches is to improve the strength property of papers. Depending upon the penetration depth of size press starch, different effects on paper property will result. This difference in penetration tendency of starches gave differing improvement of tensile strength and stiffness. As shown in **Table II**, handsheets surface sized with oxidized starch showed greater tensile strength, while those surface sized with cationic starch gave better stiffness.

Effect of starch type on broke recycling

Self-retention of size press starch onto fiber surface is critical for reducing white water contamination of the papermaking process and decreasing the loss of raw materials to drain. The amount of starch desorbed when size-pressed paper was recycled was determined using surface sized handsheets. Results showed that around 80% of cationic starch remained adsorbed on the papermaking fiber while 35% of oxidized starch retained adsorbed after repulping. It has been shown that the adsorption of cationic starch onto cellulosic fibers and coating clay is to a great extent governed by the electrostatic interaction between the charged fiber material and cationic starch molecules [10-12]. This high affinity of cationic starches for the cellulosic fibers causes a low BOD load in the effluent water.

Filler retention decreased around 15% when surface-sized paper with oxidized starch was recycled because anionic starch decreased retention efficiency of the cationic PAM (**Fig. 6**). Similar results were obtained for fines retention indicating that when cationic starch was used for surface sizing less amount of retention aid will be needed for retention. Substitution of oxidized starch with cationic starch for surface sizing reduced the cationic demand of white water filtrate more than 25%. COD of white water also decreased more than 50 ppm when cationic size press starch was used (**Fig. 7**).



5. CLSM images of the cross section of paper surface sized with oxidized starch (left) or cationic starch (right).

	CATIONIC STARCH-I	AMMONIUM PERSULFATE STARCH	OXIDIZED STARCH
Tensile index (Nm/g)	49.9 ± 0.7	51.5 ± 1.0	51.8 ± 1.1
Stiffness (mNm)	0.503 ± 0.025	0.443 ± 0.027	0.411 ± 0.022

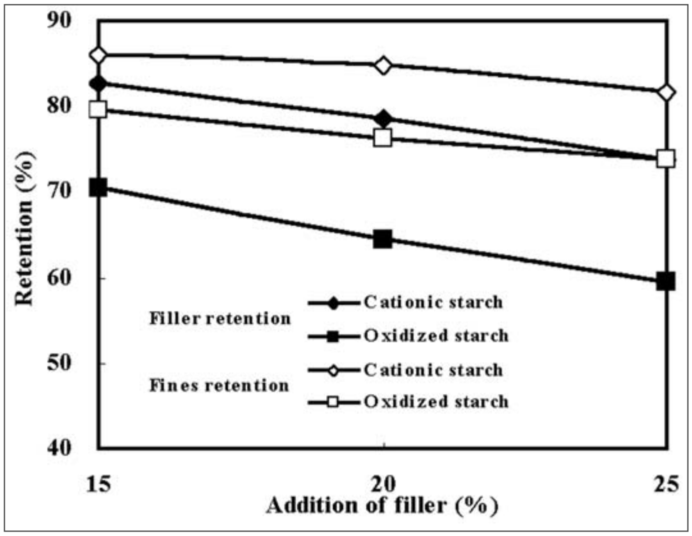
II. Effect of surface sizing starches on tensile strength and stiffness (with test results reported as mean values for 12 measurements along with 95% confidence intervals)

The detrimental impact of oxidized starch desorbed from the recycled papers increases exponentially as the white water closure increases [13]. Furthermore, the temperature of the papermaking system increases with the increase of the closure level and this enhances the ratio of starch desorption from the broke [6]. Thus, the advantages of using cationic starches increases when the level of papermaking system closure is increased.

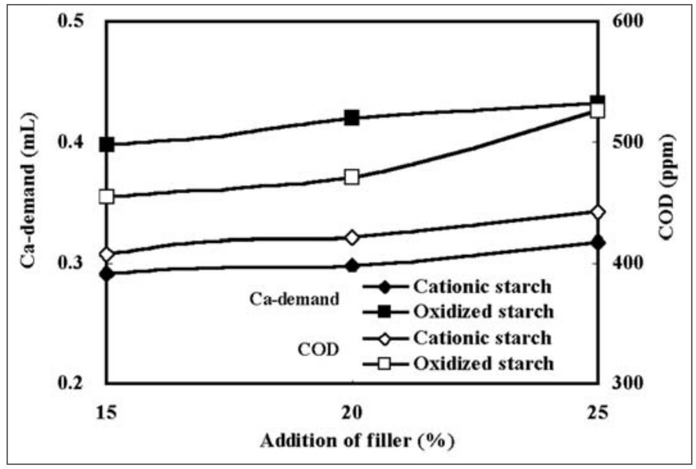
Effect of starch types on pigment coating

The interaction between coating color component and base stock is critical for the structural formation of coating layers. The concept of improving fiber coverage using a cationic coating color that rapidly immobilizes through interpolymeric complex formation with the anionic paper surface has been exploited. Other studies obtained better coverage and improvement of optical and printing properties at a lower blade pressure and coat weight [8]. Implementation of this concept is still delayed, however, because expensive coating ingredients are required. Research suggests that electrostatic interactions between the paper substrate and coating color which promote destabilization or flocculation would promote rapid immobilization and increase the porosity and many properties associated with the pore structure of the coating including opacity, brightness, ink absorption, etc [14, 15]. It is quite logical, therefore, to expect to have more structured coating with greater pore volume when cationic starch was used for surface sizing the base stock because there will be stronger electrostatic interaction between the cationic paper surface and anionic coating components.

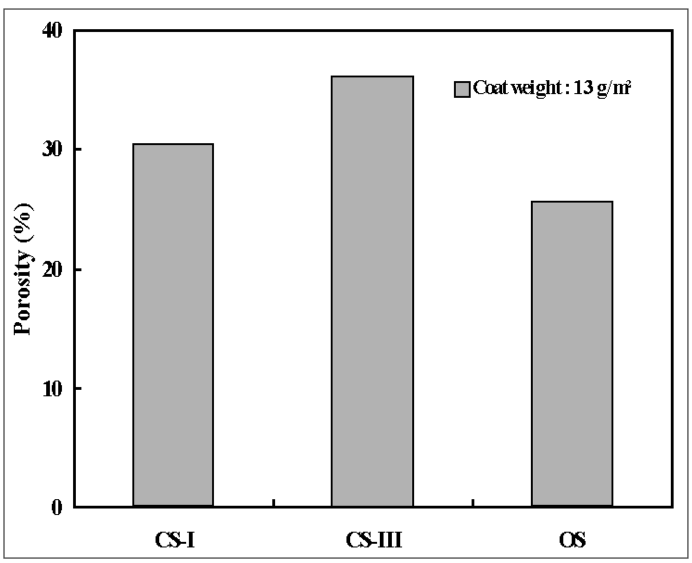
To investigate this hypothesis, we coated transparent plastic films with either cationic starch or oxidized starch. We applied pigment coating onto these films to reach 13 g/m^2 of coat weight. Our results showed that the coating layer formed on the substrate surface sized with a cationic starch (CS-III) gave the highest pore volume. The coating formed on the substrate surface sized with oxidized starch showed the lowest pore volume (**Fig. 8**). Higher brightness



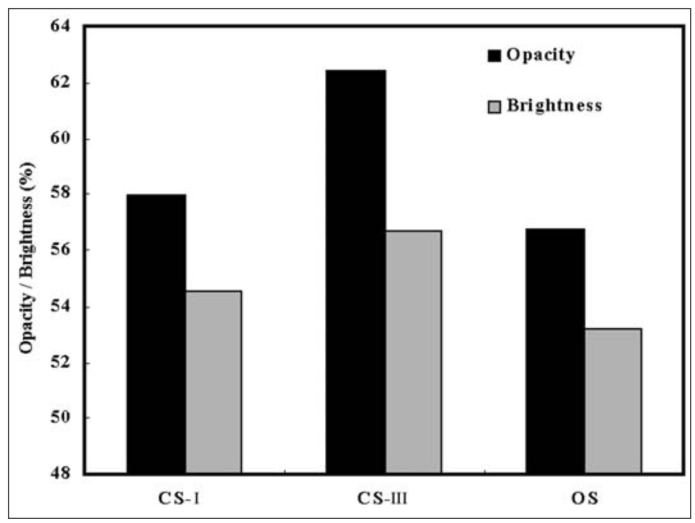
6. Filler and fines retention vs. filler addition for cationic and oxidized starches.



7. Cationic demand and COD of the DDJ filtrates.



8. Effects of starches on coating porosity.



9. Effect of starches on opacity and brightness of coating formed on plastic film.

and opacity also were obtained for the coatings formed on the plastic film surface sized with CS-III (**Fig. 9**). The fact that CS-III–with a higher degree of substitution–gave the highest porosity indicates that stronger interaction is beneficial for creating more pore volume in the coating.

Mill experiences

We tested the benefits of using cationic starch at the size press on PM 23 (which was equipped with a film transfer size press) of Hansol Paper Company for 34 days from August 31 to October 3, 2000. This machine produces coating base stock in alkaline condition. Its average machine speed during the trial period was about 1150 m/min. The average furnish composition during this trial was 75% hardwood bleached kraft pulp and 25% other pulps consisting of softwood bleached kraft pulp and BCTMP. The degree of substitution of the cationic starch was 0.015, and the viscosity of 10% starch solution ranged from 10-15 cPs.

Properties of surface-sized paper before and after pigment coating were determined, and the average of 10 measurements was reported. Paper properties evaluated were ash content, starch pickup, internal bond strength, stiffness, and gloss. Ash content was determined after ashing the paper at 525°C. Picking resistance, trapping and ink setting were determined with RI printability tester. Same inks and methods as used to carry out the RI printability tests for the laboratory prepared sheets were employed. Starch content, COD and conductivity for the stock samples taken from the headbox and silo were monitored. Enzymatic test method was used to determine the starch content [16].

Table III shows representative results obtained from the mill trial. As seen here coating base papers with better internal bond strength and picking resistance were obtained when ammonium persulfate starch was substituted with cationic starch at the size press. When these papers were coated, improvement in ink set and trapping were obtained; gloss and picking resistance were about the same. This was very promising since the paper size pressed with cationic starch contained more ash and had slightly less amount of size press starch than the paper sized pressed with ammonium persulfate starch.

When the ash contents of papers were maintained more or less constant, even greater improvement in internal bond strength, stiffness, and picking resistance of base paper were obtained when we used cationic size press starch. In addition, substantial increases in print gloss, stiffness and picking resistance for coated papers were achieved when cationic starch was used, as shown in **Table IV**. This suggests that cost reductions are possible by using more fillers when cationic size press starch is used. It was surprising to see that cationic size press starch improved internal bond strength of base paper. Oxidized size press starch tends to penetrate deeply into paper structure, while cationic starch tends to stay on the surface (Fig. 5). The results obtained from laboratory handsheets showed that oxidized starch gave better internal bond strength as well [17]. We attributed the greater internal bond strength obtained when cationic starch was used at the size press to the greater amount of cationic starch left on fiber surface after repulping due to its lower extractable character. That allowed the starch remaining on fiber surfaces to act as dry strength agents.

Improvement of strength properties achieved using cationic starch at the size press allows for an increase in filler content and resulting cost reductions [6]. Consequently, we examined the possibility of increasing filler levels. During this mill trial the ash content of the coating base paper at a basis weight of 63 g/m^2 increased from 15.4%-16.6% to 16.9%-18.9%. The results indicated that internal bond strength increased more than 30%, and stiffness and picking resistance remained rather constant although the ash content increased as mush as 2.0% (**Table V**).

To examine the desorbing tendency of size press starches into white water, we determined the starch contents in the headbox and silo. As shown in **Table VI**, substitution of ammonium persulfate starch with cationic starch resulted in the

PROPERTY	AMMONIUM PERSULFATE STARCH	CATIONIC STARCH
Base paper property		
Surface sizing starch (%)	5.15	5.03
Ash content (%)	14.53	15.50
Scott internal bond strength (ft·lb×10^{-3})	165	250
RI picking resistance	4.1/4.3	4.3/4.5
Coated paper property		
Paper gloss (%)	72.0	72.5
Printing gloss (%)	78.3	78.5
RI picking resistance	4.0/4.3	4.1/4.3
Ink set	4.1/3.8	4.5/4.3
Trapping	4.0/3.8	4.3/4.2

III. Properties of coating base stock (74 g/m^2) and coated paper (104 g/m^2)

PROPERTY	AMMONIUM PERSULFATE STARCH	CATIONIC STARCH
Base paper property		
Surface sizing starch (%)	4.73	5.03
Ash content (%)	15.41	15.50
Scott internal bond strength (ft·lb×10^{-3})	170	250
Stiffness (mNm)	72.0	74.5
RI picking resistance	3.8/4.0	4.3/4.5
Coated paper property		
Paper gloss (%)	40.5	41.5
Printing gloss (%)	67.8	70.5
Stiffness	63.0	71.8
RI picking resistance	3.9/4.2	4.3/4.3
Trapping	4.0/4.2	4.0/4.2

IV. Properties of coating base stock (63 g/m^2) and coated paper (80 g/m^2)

	AMMONIUM PERSULFATE STARCH		CATIONIC STARCH				
Ash content (%)	15.4	16.6	18.8	18.9	16.9	17.4	17.7
Internal bond (ft·lb×10^{-3})	170	175	240	235	260	240	250
Stiffness (mNm)	72.0	67.3	68.5	66.0	70.3	68.8	67.0
Picking resistance	4.0	3.9	4.2	-	4.1	-	4.0

V. Comparative study for increasing ash content of a coating base paper

	AMMONIUM PERSULFATE STARCH	CATIONIC STARCH	REDUCTION (%)
Starch content (ppm)			
Headbox	680	280	59
Silo	650	250	62
COD (ppm)			
Headbox	1400	1000	29
Silo	1380	980	29
Conductivity (mS/cm)			
Headbox	1.4	1.2	14
Silo	1.4	1.2	14

VI. Wet end analysis results

reduction of starch content by about 60%, indicating that substantially greater amounts of cationic size press starches remained on fiber surfaces during broke recycling. This would be very effective in reducing anionic trash in the process water and in utilizing raw materials more effectively. COD and conductivity in the headbox and silo decreased when we used cationic starch. Reductions in COD will result in effluent load reductions, and lower conductivity will improve the effectiveness of many polyelectrolytes.

We also measured ash content at the storage tank of the size press during the mill trial. We found that average ash content of the size press starch decreased from 2.16% to 1.52% after substituting ammonium persulfate starch with cationic starch. The ash contents of cationic starch and ammonium persulfate starch were 0.8% and 0.1%, respectively. This indicates that the reduction of ash content in the size press starch is not due to starches used. Since the ash in the starch tank derives from the fillers detached from the paper surface, this level is closely related to the runnability of the film transfer size press equipped with rod bars.

CONCLUSIONS

Use of oxidized starches at the size press causes diverse problems in broke recycling and reduces opacity, brightness and printing properties because of their anionic character. To solve these problems associated with oxidized starches cationic starches were prepared and the effects of the type of cationic starches on paper properties and papermaking processes were investigated.

Results showed that cationic starches have pronounced effects in improving opacity, brightness, print gloss, and ink density of paper due to its better holdout property. Effectiveness in surface holdout of cationic starches was verified by CLSM. Adsorption ratio of the cationic starch onto fiber was 80%, which was substantially larger than that of oxidized starch. This decreased COD load in white water by more than 50 ppm and increased fines retention and strengths of handsheets. Base papers surface sized with cationic starch also gave better opacity and brightness of coated paper.

When cationic starch was used as size press starch for a film transfer size press the starch contents in headbox and white water silo were decreased by 59%-62% and 29%, respectively. Improvement in internal bond strength, stiffness, and picking resistance of paper was obtained when ammonium persulfate starch was substituted with cationic starch and this allowed the room to use more fillers. **TJ**

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