
Klaus Doelle, Anh T. Le, Dave C. Bailey, Matthew Zelie, Tim McNaney, Dylan M. Salpeter, Joseph Piazza, Matthew T. Wainwright, Rachel J. Klein, Stephen P. Tramposch, Kyle Dausman, Autumn R. Elniski, Candace Guilford, Andrew J. Murphy, Thomas J. Rimmer, Nicholas Rozanski, Christopher G. Strezywilk, Seth P. Walters, and Yipeng Yan

Today recycled paper is used to reduce the environmental impact of paper production. Starch as a dry strength additive improves the properties of recycled paper. Starch in combination with Poly-lactic Acid (PLA), a product from the biorefinery process, has the potential to act as a promising strength additive to further improve paper strength. In this study, PLA with molecular weight of 20,000, recycled pulp (OCC) with kappa number of 56.6, and cationic starch containing 0.43% nitrogen content were employed. Three machine conditions were run: Condition 1 – only pulp; condition 2 - pulp plus 1% starch; condition 3 - pulp plus 0.9% starch and 0.1% PLA. Selected paper properties were tested in accordance to TAPPI standards. The results showed that tensile index in the cross direction (CD) increased about 26% for the paper made in condition 3 compared to condition 2. Tear strength of paper in condition 3 also showed a slight increase in the machine direction (MD), while it remained the same in CD, compared to the condition 2. There was no improvement seen for air and water resistance. Therefore, this study suggests that a blend of PLA – starch represents a promising approach in improving the strength properties of recycled paper.

Keywords: PLA; Recycled pulp (OCC); Cationic starch; Paper properties; Strength, Paper machine.

Contact information: Department of Paper and Bioprocess Engineering, State University of New York College of Science and Forestry, 1 Forestry Drive, Syracuse, NY 13210, USA;
*Corresponding author: kdoellle@esf.edu

INTRODUCTION

At the present time, papermakers are focusing on how to improve the strength of paper. This can be measured by tensile strength, bursting strength, or internal bonding strength. There are various ways to increase the strength of paper such as using refining, wet pressure, or additives. However, use of mechanical actions such as refining could bring about the reduction in opacity, brightness, dimensional stability, or porosity (Scott 1996). Therefore, the use of additives is very common and has been employed in most paper mills. Starch is used today as a dry strength additive in the paper industry. Starches can significantly improve the mechanical properties of paper, such as tensile strength. Also, starches are used as retention aid, surface sizing agent, coating binder, and adhesive in corrugated board and other converting operations (Biermann 1996).

Although they can provide the excellent contributions to the strength and other properties of paper, the use of starches can also lead to several problems in
the papermaking process. One of the current major problems when using starches is the loss of starch to the white water system. Starches are water soluble, hydrophilic polymers (Neimo 1999). When the amount of starch added to the system is high, the retention of starch on the paper web decreases and then starch is lost to the white water system due to its hydrophilic properties. The loss of starches results in increased production costs, and in the long term of operation, it can cause microbiological problems that negatively affect the runnability of machines as well as the quality of paper. Therefore, it is necessary to study new additives to use together with starch to further increase the strength of paper, reduce the consumption of starch, and reduce the microbiological problems.

In an attempt to fulfill these purposes, poly-lactic acid (PLA) had been introduced to the wet end system. PLA has been found to be a potential alternative for starch, and it is also a biodegradable product. Furthermore, PLA can be produced from natural resources like corn, but also woody biomass. Recently PLA has been shown in a laboratory study to act as a strength additive (Gong et al. 2012; Hasan et al. 2010). It was reported that the blend of PLA with cationic starch as wet end additives improved the tensile strength of paper. According to Gong, several virgin pulps were used, including Norway spruce thermo-mechanical pulp (TMP), hardwood unbleached kraft pulp, and softwood bleached kraft pulp. Two cationic starches having different nitrogen contents, and PLA with molecule weight of 20,000 to 30,000 were employed.

In this paper machine run, recycled pulp was used instead of the virgin pulps. This was done because recycled paper is broadly used today for environmental reasons. Furthermore, it has been shown that some paper properties from recycled pulp could be comparable to paper properties from virgin pulp. For example, after several times of recycling, mechanical pulp still gives better density, better burst strength and better tensile strength compared to the never-dried mechanical pulp (Smook 2002). However, for chemical pulp, important paper properties such as burst and tensile strength decrease when the number of recycles increases. The reason for the decrease in strength is due to the loss of bonding potential between fibers after they were dried in the paper drying process. One of the methods to recover the strength of paper is to use additives like modified starch.

In this present paper, we report the results of selected properties of paper made from the three machine conditions.

EXPERIMENTAL

Materials

Pulp used for this paper machine run was Old Corrugated Container (OCC) recycled pulp provided by a paper mill in central New York, possessing a kappa number of 56.6, and having an initial Canadian Standard Freeness (CSF) of 559 mL. The pulp was then refined to a final CSF of 281 mL.

Starch used in this run was cationic starch. Its catalog number, provided by a starch company in the United States. This starch has a nitrogen content of 0.43%, which is considered to be a high cationic charge starch.

PLA was purchased from a company in the United States with a molecular weight of 20,000 to 30,000 Daltons.
Methods

*TAPPI methods*

Kappa number of the recycled pulp was measured in accordance with T236 om-06, “Kappa number of pulp”. The CSF was measured by T227 om-09 “Freeness of pulp (Canadian standard method)”. The grammage was determined by T410 om-08 “Grammage of Paper and Paperboard (weight per unit area)”. The thickness was measured by T411 om-10 “Thickness (caliper) of paper, paperboard, and combined board”. Tensile strength was performed following T494 om-06, “Tensile properties of paper and paperboard (using constant rate of elongation apparatus)”. The tear strength was done by following the T414 om-12, “Internal tearing resistance of paper (Elmendorf-type method)”. Sheffield porosity and smoothness were tested using T547 om-07 “Air permanence of paper and paperboard (Sheffield method)” and T538 om-08, “Roughness of paper and paperboard (Sheffield method)”, respectively. The Cobb values were obtained by following T441 om-09, “Water absorptiveness of sized (non-bibulous) paper, paperboard, and corrugated fibreboard (Cobb test)”.

*PLA treatment*

PLA is a hydrophobic polymer and does not readily dissolve in water; it was dissolved in a minimal amount of acetone. In this run, 23 mL of acetone was used to dissolve 4g of PLA, under constant stirring speed in a small beaker, and for about five minutes.

*Starch preparation*

Starch was cooked in a batch at about 2.5 % solids. Raw starch and water were placed in a metal beaker, which was then placed on a heating and stirring device. It was then cooked at 95 to 98 °C for 30 minutes, and at a constant stirring speed.

*Mixing of Starch – PLA*

After PLA was dissolved in acetone, it was mixed with the cooked starch and they together formed a clear and stable solution. Then the Starch-PLA mixture was added to the pulp slurry and agitated for about 5 minutes to get an even mixture of pulp, starch and PLA.

*Paper Machine Conditions*

The 12 inches pilot plant papermachine at the State University of New York College of Environmental Science and Forestry (SUNY-ESF) was used for this run. Details for the three conditions are described below:

Condition 1 (blank – pulp only, no starch or PLA): The wire speed was 10 ft/min. The nip pressure was 20 psig for the 1st press, and 25 psig for the 2nd press. Nineteen (19) dryer cans were used with the drying temperature range from 200 to 330°F. This condition was not calendered.

Condition 2 (pulp + 1% cationic starch, no PLA): The wire speed was 10 ft/min. The pressure nip was 20 psi for the 1st press, and 25 psi for the 2nd press. Nineteen (19) dryer cans were employed with the drying temperature range from 210 to 400°F. This condition was calendered at several different pressures (10, 20, 30, 40, and 50 psi).

Condition 3 (pulp + 0.9% cationic starch + 0.1% PLA): The wire speed was 10 ft/min. The 1st press pressure was 20 psig, and was 25 psig for the 2nd press.
pressure. The drying temperatures range from 210 to 400°F (19 dryer cans). This condition was calendered at several different pressures (10, 20, 30, and 50 psi).

RESULTS AND DISCUSSION

Non-Calendered Conditions

Figure 1 shows the tensile index (TI) in machine direction (MD) and cross direction (CD) of paper made from the three non-calendered conditions.

![Tensile Index, Nm/g](image)

**Fig. 1.** TI in MD and CD of the three conditions without calendering

It can be clearly seen from Fig. 1 that the TI in CD increased from 17.32 to 19.91 Nm/g (about 15% increase) from condition 1 to condition 2, which was treated with 1% starch. As expected, TI in CD of condition 3, which was treated with 0.9% starch plus 0.1% PLA, increased significantly to 25.08 Nm/g (about 26%) compared to that of condition 2. This result suggests that with the presence of starch and PLA, the bonding degree between fibers increases, resulting in a strength increase of paper. Also, with the presence of the hydrophobic PLA, the association of PLA and the lignin-rich pulp becomes stronger, leading to a higher retention of starch and PLA in the paper structure, hence resulting in the increase in paper strength. Unlike the TI in CD, TI in MD of condition 2 (1% starch) reached the highest value at 58.87 Nm/g. It increased about 13 units (about 28%) compared to that of condition 1 at 45.90 Nm/g, which was not treated with any starch or PLA. Unexpectedly, the TI of condition 3 in MD at 48.33 Nm/g is lower than that of condition 2. However, the TI of condition 3 in MD was greater (about 5.3%) than that of condition 1.

Figure 2 shows the tear index of the three non-calendered conditions. It is apparent that tear index in CD is higher than that in MD because fibers tends to orient more in MD than CD. In MD, tear index slightly increased for condition 2 (10.22 mN*m^2/g) and condition 3 (10.86 mN*m^2/g) compared to that of condition 1 at 9.63 mN*m^2/g. For tear index in CD, conditions 2 and 3 were seen to be almost the same (12.35 vs 12.18 mN*m^2/g), and were also greater than condition 1 at 11.63 mN*m^2/g.
These results indicate that in the presence of starch and PLA, tear strength of paper slightly increased compared to the blank.

![Tear index, mN·m²/g](image)

**Fig. 2.** Tear index in MD and CD of the three conditions without calendering

Figure 3 shows the porosity, Cobb value, thickness and grammage of the papers made from the three non-calendered conditions.

![Porosity, Cobb value, grammage and thickness](image)

**Fig. 3.** Porosity, Cobb value, grammage and thickness of the three conditions without calendering

According to Fig.3, the lowest Cobb value was seen for condition 2 at 76.2 g of water/m², and was lower than that (98.4 g of water/m²) of condition 1 by about 22.6 %. This result indicates that with the presence of 1% starch, less water penetrated the paper structure within the 120s test time. Unexpectedly, when 0.1% PLA was added together with 0.9% starch, the amount of water staying in the paper was 88.20 g/m².
which was higher than that of the paper made with 1% starch. The results suggest that even with the presence of the hydrophobic PLA, the water resistance of paper was not improved in this paper machine run.

The thickness and grammage results were also presented in Fig.3 to show the correlation between these properties and the porosity of paper. It can be seen that the highest porosity was recorded for condition 2 at 2262 sccm. This porosity (air flow rate) corresponds to the lowest grammage (50.65 gsm) and the thinnest paper (161 µm). The porosity of conditions 1 and 3 were almost the same (1594 vs 1603 sccm), although the thickness of paper of condition 1 was about 9 µm (175 vs 184 µm) lower than that of condition 3. Therefore, in this paper machine run, these results indicate that the presence of starch and PLA caused an increase in the porosity of the paper. This result is unexpected because it is believed that the presence of starch would fill pores in the paper structure.

**Calendered Conditions**

*Condition 2 - 1% starch*

Figure 4 shows the effects of calender nip pressure on thickness, porosity and smoothness of paper made in condition 2.

As can be seen from Fig.4 when the nip pressure increased from 10 to 50 psi, the thickness of the paper decreased from 134 µm to 114 µm (about 15%). For smoothness, it is also clear that the air flow rate decreased from 2792 sccm at 10 psi to 2442 sccm at 50 psi (About 12.5% decrease). These results indicate that the paper surfaces became smoother with the increase of nip pressure. For the most part, porosity results also followed the expected trend, decreasing from 2093 sccm to 2021 sccm as the calender pressure increased from 10 to 50 psi. This is due to decrease in pore volume caused by the compaction of the sheet as calendering pressure increases. One exception seen here is the porosity at the nip pressure of 20 psi (1948 sccm).
Condition 3 – 0.9% Starch + 0.1% PLA

Figure 5 shows the effects of calender nip pressure on thickness, porosity and smoothness of paper made in condition 3.

As shown above, thickness was reduced from 177 µm to 110 µm (About 37.8% reduction) with the increase of nip pressure from 10 psi to 50 psi. Porosity was seen to decrease from 1373 sccm at 10 psi to 1097 sccm at 50 psi (About 20% reduction). For the smoothness, except for the value of 1422 at 30 psi, airflow rate was seen to decrease from 2701 sccm at 10 psi to 2055 sccm at 50 psi (About 24% reduction). The results again confirm that paper smoothness increases with an increase in calendar nip pressure.

CONCLUSIONS

Laboratory paper machine runs were performed to evaluate the effects of the use of Poly-lactic Acid in combination with starch in the wet end system, on selected properties of recycled paper. Results are summarized below:

1. Tensile index in CD increases about 26% for the paper made with 0.9% starch plus 0.1% PLA compared to the paper made 1% starch. This result suggests that with the combination of PLA and starch, the bonding degree between fibers increases, resulting in an increase in the CD strength of the paper. Also, with the presence of the hydrophobic PLA, it is believed that the association of PLA and the lignin-rich pulp becomes stronger, leading to a higher retention of starch and PLA in the paper structure, hence resulting in an increase in paper strength.

2. With the presence of PLA, tear strength in MD slightly increased compared to that of paper made with only 1% starch. Tear strength in CD of the paper made with 1% starch, when compared to paper with 0.9% starch plus 0.1% PLA was almost the same, and these were higher than that of paper made with the pulp only.
3. The Cobb values show that the water resistance of paper made with 0.9% starch plus 0.1% PLA was not improved compared to that of paper made with 1% starch.

4. The Porosity values show that the air resistance of paper made with 0.9% starch plus 0.1% PLA was not improved compared to that of paper made with 1% starch.

In this study, we also used the calendar for condition 2 and condition 3 to see the effects of calendar nip pressure, when PLA was used, on the thickness, smoothness, and porosity of paper. The results indicate that with an increase in nip pressure, thickness and porosity decrease, and smoothness increases.

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