IMPROVEMENT OF RECYCLED PAPER'S PROPERTIES FOR THE PRODUCTION OF BRAILLE PAPER BY IMPREGNATION WITH LOW GRADE CELLULOSE ACETATE: OPTIMIZATION USING RESPONSE SURFACE METHODOLOGY (RSM)

Mei Chong Soo, Wan Rosli Wan Daud, and Cheu Peng Leh*

Paper dust is a kind of cellulosic waste that is generated by converting operations in paper mills. It was derived to a low-grade cellulose acetate in this study. Papers made from recycled fiber were then impregnated with the resultant cellulose acetate. Effects of impregnation conditions on the paper properties were statistically investigated by employing central composite design (CCD) based response surface methodology (RSM). Four response variables, namely density, burst index, smoothness, and rate of surface wettability were analyzed. Polynomial estimation model of each response was developed as functions of three independent variables, which are pressing temperature (T), dipping time (D), and concentration of cellulose acetate (C). The paper which was impregnated based on the calculated optimum condition (T: 163 °C, D: 2.8 minutes, and C: 2.7 percent), possessed a density of 0.5450 g/cm³, rate of surface wettability of 0.012°/s, burst index of 2.84 kPa m²/g, and paper smoothness of 475 mL/min. There was no significant difference between the experimental values and the predicted values calculated from estimation models. The cellulose acetate impregnated Braille papers made from recycled fibre was found to have better properties than those of commercial Braille paper in terms of rate of surface wettability and burst index.

Keywords: Braille paper; Cellulose acetate impregnation; Recycled paper; Response surface methodology

Contact information: Bio-resource, Paper and Coating Divisions, School of Industrial Technology, Universiti Sains Malaysia, 11800 Minden, Penang, Malaysia * Corresponding author: cpleh@usm.my

INTRODUCTION

Cellulose acetate is a kind of thermoplastic, and it is one of the most important and widely used cellulose derivatives (Ibrahem *et al.* 1983). Its applications include woven fabrics, braids, medical gauze, women's lining, cigarette tows, and dialysis membrane. Besides, cellulose acetate could be blended with other fibres to make combination yarns (Law 2004).

The application of cellulose acetate in papermaking had been reported since 1977, when Litzinger (1977) patented an extrusion process of making cellulose acetate fibers, which was suitable to be used in conventional papermaking methods (Rustemeyer 2004). In the same year, Keith (1977) produced a kind of cellulose acetate fiber for papermaking applications by precipitation of cellulose acetate from a dope under high shear conditions. In addition, a kind of paper with an inner core of cellulose acetate has been invented by Frederick *et al.* (1997) and Mitchell *et al.* (2000), where the former aimed to enhance the

thermoplastic properties of the paper. On the other hand, the latter intended to overcome the disadvantage of paper such as permeability by water and rigidity of the paper, while it still maintained the biodegradable properties of the paper products. Lamination of several layers of cellulose acetate film with paper under high pressure and heat was invented to improve the post-formability of the laminates of décor sheets (Cox *et al.* 1997).

The chemical and enzymatic degradation reactions of cellulose acetate are widely dependent on the degree of substitution (DS) (Sjöholm 2004). The main affecting parameters of DS are the method of esterification and the properties of the raw material used. Dissolving pulp, also known as chemical cellulose, is the major raw material for the production of high-quality cellulose acetate. This pulp is categorized by its high alphacellulose content (greater than 95 percent), low hemicellulose content (less than 4 percent), and traces of lignin and ash content.

Recently, the production of cellulose acetate from lower grade pulp such as secondary fiber, agrowaste fiber, and non-wood biomass pulp, has been reported (Filho *et al.* 2008). However, the cellulose acetate produced showed relatively poorer quality in terms of transparency, thermal stability, filterability, and solubility. These inferior properties were due to the presence of impurities in the materials, which included lignin, hemicellulose, and low molecular weight cellulose. Nevertheless, the resultant cellulose acetate with a high degree of substitution (DS) could be obtained when applying a longer acetylation time. The potential applications of this lower-grade cellulose acetate are numerous, including: a variety of coating applications, pressure-sensitive tapes, wood-sealers, packaging, and paper and paperboard for food contact applications (Eastman 2006).

In the world of blind or visual-impaired people, listening and touching are the main ways to gain knowledge. Although in this advanced age, much information can be accessed through electronic devices, the basic communication medium, Braille-printed materials, is still very important to the group of people, especially to those from developing countries, who may lack electronic Braille tools.

The Braille system is a type of tactile symbol based on the Braille cell, which is composed of one to six dots in a specific position (Rex *et al.* 1995). Braille dots are produced by using a special typewriter known as an embosser. To ensure that the Braille dots produced have sufficient stiffness and height for effective reading, Braille paper basically requires a relatively high grammage of 120 to 140 gsm and an apparent density of more than 0.75 g/cm³. In order to meet this requirement, high density and fully bleached virgin paper, which principally has the better inter-fibre bonding ability (Casey 1981), is normally used as raw material. However, to achieve environmental sustainability, utilization of recycled paper for the production of Braille paper is recommended. Nevertheless, some modifications are required during the papermaking process to attain the desired properties of Braille paper. One of the methods to improve paper properties is via a simple impregnation or coating method by dipping paper sheets in a polymer solution (Ibrahem *et al.* 1983).

Paper dust is a type of cellulose waste generated in the course of converting operations in a paper mill and cannot be recycled for papermaking, as it has rather short fiber length. Although cellulose is the most abundant natural resource, it is valuable and should be fully utilized. In order to retrieve and to maximize the resource value, paper dust was derivitized to form a low-grade cellulose acetate in our previous study (Soo *et*

al. 2010; Loo *et al.* 2012). Recycled papers were then impregnated with the resultant cellulose acetate with the aim to improve the properties of the papers, so that it was suitable to be used for Braille embossing. Based on the analysis of a half 2-level factorial (2k-2) design of three independent variables—dipping time, pressing time, and pressing temperature, it was found that the dipping time was the most influential variable, followed by the pressing temperature. Nevertheless, the pressing time showed an insignificant effect on all the responses.

In this study, the effect of the impregnation process variables (the pressing temperature (T) and dipping time (D) and concentration of cellulose acetate bath (C)) on paper properties was investigated by employing experimental design, namely central composite design (CCD) based surface response methodology (RSM). The quality of the paper was evaluated based on four response variables: density, burst index, smoothness, and rate of surface wettability. The estimated models of the four responses generated were used to identify the optimum process conditions.

MATERIALS AND METHODS

Material

Secondary paper dust, which was used to produce cellulose acetate, was provided by Massive Paper Mill Sdn Bhd, Kedah, Malaysia, a corrugated carton box-converting mill; whereas the secondary fiber (kraftliner board) was used to produce handsheets and was provided by Muda Paper Mill Sdn Bhd, Penang, Malaysia.

Preparation of Cellulose Acetate

Five grams of paper dust was dipped into 100 mL of deionized water in a 250 mL beaker for 10 minutes. Next, the deionized water was filtered out by using a G3 sintered crucible, and the paper dust was then dipped into 100 mL acetic acid glacial in a 250 mL beaker for another 10 minutes. After that, the paper dust was filtered and dipped into another 100 mL of fresh glacial acetic acid to activate the pulp.

Acetic acid glacial, 90 mL, and 0.5 mL of concentrated sulfuric acid were poured into a 250 mL conical flask with a stopper. Then the flask was kept in a water bath at 25 °C. The paper dust, which had been activated earlier, was added into the particular flask and shaken for 1 minute. After that, 25 mL of acetic anhydride was added into the flask and shaken for another 1 minute. The mixture was kept in the water bath at 25 °C for 60 minutes. Subsequently, 12.5 mL of glacial acetic acid – water mixture (acetic acid: water = 19:1 in volume) was added into the flask and the cellulose solution was stirred thoroughly for 30 minutes at 25 °C.

Next, the acetylated cellulose mixture was poured slowly into a 2.5-L beaker containing 1.2 to 1.4 L of deionized water. If the droplet of the cellulose mixture looked like gel and did not disperse in the deionized water, the acetic acid-water mixture was added into the cellulose mixture for dilution. On the other hand, if the droplet looked like an emulsion, the speed of the stirrer was slowed down. After completion, the precipitated acetylated cellulose (cellulose acetate, CA) was squeezed and dispersed into 1.2 to 1.4 L deionized water in a 2-L beaker. The precipitated CA was then washed several times. A few drops of phenolphthalein were added into the beaker, and sodium carbonate was slowly added into it until the mixture became a pinkish color. Finally, the mixture was

filtered out and washed with deionized water several times, followed by drying in an oven at 60 to 80 $^{\circ}$ C. The dry sample was then ground into power form by using a porcelain mortar.

Preparation of Secondary Pulp Hand Sheet

The kraft-liner (made from recycled paper) was torn into small pieces and soaked in water for no less than 6 hours before handsheet making. The procedure of handsheet making was according to TAPPI method T-205sp-02 with the minor modification, where instead of 60 gsm, handsheets with 80 gsm were produced.

Impregnation of Hand Sheet with Dissolved Cellulose Acetate

A secondary pulp handsheet was dipped into a bath containing dissolved cellulose acetate with different concentrations (1.32%, 1.8%, 2.5%, 3.2%, 3.68%) in acetone. The handsheet was removed from the bath after an appropriate time (1.41 minutes, 2 minutes, 7 minutes, 12 minutes, and 15.41 minutes) and dried in an oven at 80 °C for 5 to 10 minutes. Then, the handsheet was pressed under a hot press machine at an appropriate temperature (72.73 °C, 100 °C, 140 °C, 180 °C, and 207 °C) for 2 minutes at a pressure of 100 psi.

Physical Properties Testing

The properties of the handsheets were tested according to TAPPI methods T403 om-2 to find the burst index, T411 om-97 to measure paper thickness, T458 om-84 to evaluate the surface wettability of paper (angle of contact method), and using a Bendtsen smoothness tester (SCAN-P 84:02), which is one type of air-leak instrument, to determine papers' smoothness/roughness (Casey 1981). The test value units reported in mL/min represent the volume of the air passing between the paper surface and the flat polished glass surface that has an area of 10 cm² in minutes. As the smoother paper is quantified with lower test value, thus, this test could be known as a roughness test. The percentage of cellulose acetate impregnated in paper was also calculated by dividing the different weight of the impregnated paper and the non-impregnated paper with the weight of non-impregnated paper, all multiplied by 100 percent. The degree of substitution (DS) of cellulose acetate was analyzed according to ASTM method D871-96 (2004).

Experimental Design

A central composite design (CCD) of the Response Surface Method (RSM) was used to analyze four response variables (paper density, the rate of surface wettability of paper, smoothness, and burst index) statistically toward three independent variables of CA impregnation (the dipping time, pressing temperature, and concentration of dipping bath) with the assistance of computer software (Expert-Design® by Stat-Ease, Inc. USA). This design usually works well for process optimization. Table 1 shows the 18 experimental conditions calculated by the software. The code values of the independent variables were calculated through Equations 1 to 3. The three center points incorporate with fractional factorial designs were aimed to estimate of pure error, as well as the test for curvature.

$T_{\rm code}$	=	(Pressing Temperature – 140) /100	(1)
$D_{\rm code}$	=	(Dipping time – 7) / 2	(2)

$$C_{\text{code}} = (Concentration of Cellulose Acetate - 2.5) / 1.8$$
 (3)

CCD was used since it makes it possible to estimate the coefficient of a quadratic model as shown by the following equation,

$$y = \theta_0 + \theta_1 x_1 + \theta_2 x_2 + \theta_3 x_3 + \theta_4 x_4 + \theta_{11} x_1^2 + \theta_{22} x_2^2 + \theta_{33} x_3^2 + \theta_{44} x_4^2 + \theta_{12} x_1 x_2 + \theta_{13} x_1 x_3 + \theta_{14} x_1 x_4 + \theta_{23} x_2 x_3 + \theta_{24} x_2 x_4 + \theta_{34} x_3 x_4$$
(4)

where y represents response variable, β_0 is the interception coefficient, β_1 , β_2 , β_3 , and β_4 are linear terms, β_{11} , β_{22} , β_{33} , and β_{44} are quadratic terms, β_{12} , β_{34} are cross-product terms, which represent an interaction between the individual factors in that term, and x_1 , x_2 , x_3 , and x_4 are independent variables studied.

RESULTS AND DISCUSSION

Properties of Cellulose Acetate

Because the paper dust used was not homogenous, the degree of substitution (DS) of the resultant cellulose acetate was in the range of 2.0 to 2.3 with the percentage of acetyl acid between 35 and 39 percent.

Regression Analysis of Estimation Models

Table 2 presents a summary of the statistical assessment of the estimation models built from three variables, combined variables, and their corresponding significant coefficient (CE) for the four responses (density, smoothness, burst index, and rate of surface wettability). Based on the results obtained from the ANOVA statistical analysis, D, there were significant effects on all four response variables, with the value of "probability > F" less than 0.05 (Table 2) and the T showing the significance on three responses, which were density, smoothness, and rate of surface wettability. Although Tdid not show a significant effect on the burst index, it showed a significant quadratic term of T, while the variable C only showed a significant effect on the burst index. There were significant effects among the interaction of T and C on smoothness and also the rate of surface wettability. The interaction of D and C showed significant effects on the rate of surface wettability.

Analysis of Cellulose Acetate Impregnation Variables on Paper Properties

Effect on paper density

Based on Table 2, the value of CE for dipping time (D) was negative, and hence the increase of D decreased the density of the paper. This is possibly caused by the amount of CA penetrating into paper which is increased with extended D (Fig. 2). As mentioned by Baker *et al.* (1942), the presence of CA in paper will increase the distance among fibers due to hindering of the formation of hydrogen bonds. **Table 1.** Experiment Conditions Used in Coated Paper and the Result of theDensity, Smoothness, Paper Smoothness, and Rate of Surface Wettability ofBraille Paper Produced

Variable code				Variabl	е		Respo	nses	
Т	D	С	Те	Dipping	Conc.	Density	Smoothnes	Burst	Rate of
			mp.	Time	of		s/roughnes	index	surface
					dipping		S		wettability
					bath	_			
			(°C)	(min)	(%)	(g/cm ³)	(mL/min)	(kPa.	(°/s)
								m²/g)	
-1	-1	-1	100	2	1.8	0.565	214	1.91	0.095
1	-1	-1	180	2	1.8	0.577	457	1.80	0.100
-1	1	-1	100	12	1.8	0.540	323	1.94	0.010
1	1	-1	180	12	1.8	0.566	700	1.83	0.055
-1	-1	1	100	2	3.2	0.525	404	2.79	0.085
1	-1	1	180	2	3.2	0.587	460	2.02	0.010
-1	1	1	100	12	3.2	0.515	-	3.47	0.100
1	1	1	180	12	3.2	0.562	625	3.25	0.035
-1.682	0	0	73	7	2.5	0.532	288	1.77	0.100
1.682	0	0	207	7	2.5	0.581	404	1.51	0.025
0	-1.1181	0	140	1.4	2.5	0.574	300	1.70	0.075
0	1.682	0	140	15	2.5	0.556	487	3.60	0.035
0	0	-1.681	140	7	1.3	0.522	549	2.23	0.030
0	0	1.681	140	7	3.7	0.547	633	-	0.100
0	0	0	140	7	2.5	0.568	422	2.78	0.040
0	0	0	140	7	2.5	0.587	510	2.41	0.050
0	0	0	140	7	2.5	0.584	580	2.56	0.040
0	0	0	140	7	2.5	0.588	440	2.45	0.070

Table 2. Statistical Assessment of	Variables to Responses
------------------------------------	------------------------

Variable	Responses								
	Density (g/cm ³)		Smoot	Smoothness/		Burst Index		Rate of surface	
			rougl	nness	(kPa.m ² /g)		wettability (°/s)		
			(mL	/min)					
	(Eq	.5)	(Ed	q.6)	(E	q.7)	(E	q.8)	
	CE	Prob > F	CE	Prob > F	CE	Prob > F	CE	Prob > F	
Intercept	0.58	-	419.9	-	2.65	-	0.059	-	
T - Temperature	0.017	0.005	65.81	0.0138	-0.12	0.1648	-0.016	0.0080	
D - Dipping Time	-8.539	0.0430	79.25	0.0071	0.38	0.0010	-0.012	0.0381	
C – Conc. of CA	-1.227	0.7350	39.07	0.1102	0.44	0.0006	6.424	0.2163	
$ T^2_{r} $	-7.248	0.6671	-	-	-0.33	0.0023	-	-	
D^2	-	-	-	-	-	-	-	-	
$ C^2 $	-0.015	0.0013	57.87	0.0202	-	-	-	-	
TC	-	-	-67.04	0.0503	-	-	-0.024	0.0034	
TD	-	-	-	-	-	-	6.250	0.3496	
DC	-	-	-	-	0.23	0.0515	-0.021	0.0068	

Table 3. Statistical analysis of Reduce Model of Response

Source	Density	Smoothness/	Burst Index	Rate of surface
	(g/cm ³)	roughness (mL/min)	(kPa.m²/g)	wettability (°/s)
Model prob>F*	0.0009	0.0036	0.0003	0.0025
R^2 #	0.7928	0.7611	0.8507	0.7981

Table 4. Properties of Commercial White Braille Paper a	Ind Uncoated Hand
Sheet	

	Commercial White	Uncoated Hand Sheet
	Braille Paper	
Grammage, g/m ²	140	80
Density, g/cm ³	0.768	0.529
Smoothness/ roughness (mL/min)	131.25	416
Bursting index (kPa.m ² /g)	0.82	1.799
Rate of surface wettability (°/s)	0.418	All absorbed into paper after
		5 seconds

When acetone in the paper dissipated, a thin layer of CA is formed between the pulp fibers, and this acts as an additional bond between pulp fibers. This additional bond increases the distance between the fibers and thus decreases the paper density. In this study, the interaction of dipping time and temperature (*TD*) did not show a significant effect on density as obtained by the preliminary study (Soo *et al.* 2010), but the effects of *T* alone and quadratic of $C(C^2)$ showed significant effects on density.

The plot of response surface shown in Fig. 1 illustrates the effect of the dipping time, pressing temperature, and concentration of CA bath on the density of Braille paper. The response surface possesses a maximum point. The maximum point of density in the region of high temperature (180 °C) is higher than in the region of low temperature (100 °C). This may result from the fact that there is less CA impregnated into the paper when a shorter dipping time is employed. Consequently, the amount of voids between the fiber networks is less, and thus a higher density is obtained.



Fig. 1. 3D response surface plot of density as a function of concentration of cellulose acetate and temperature with a fixed dipping time at 2 minutes and 12 minutes





Effect on paper smoothness

Smoothness is a way to test the roughness of a paper and it is concerned with the surface contour of the paper. The smoothness of the Braille paper is important because it affects the touching sensation of the users. The smoothness of Braille papers produced by using secondary fibre is in the range of 214 mL/min to 700 mL/min (Table 1), while the smoothness of commercial white Braille paper is 131.25 mL/min (Table 3). This indicated that the commercial Braille paper is much smoother than the secondary fibre Braille paper.

The smoothness of the CA impregnated paper was low, possibly caused by the uneven CA distribution on the paper surface.





Based on the estimation model (Eq. 6), as Coefficient Estimates of T, D, and C are of positive value, the increase of these factors increases the value of air flow over the paper, and flat polished glass surface, or in other words, the paper smoothness is decreased. As shown in Fig. 3, the region of high temperature and dipping time resulted in the highest value of airflows through the inductor surface. In order to improve the smoothness of paper, better lab-scale impregnation and calendaring techniques have to be developed. Other than that, the smoothness may be overcome by adding a layer of coating.

Effect on paper bursting index

Burst strength is defined as the hydrostatic pressure requested to rupture a paper and also is an indicator to the toughness of paper (Casey 1981). This property is important to Braille paper for ensuring that the paper is not easy to puncture and/or further rupture during the embossing process. From the statistical result shown in Table 2, burst index is significantly affected by D, C, and T^2 . There is also an interaction between D and C. However, the T alone has insignificant effect on the bursting index.

As shown in Fig. 4, when the *D* is increased, the burst index increases dramatically in the region of high *C*, but it only increases with a small extent in the region of low C. As mentioned earlier, the increase of *D* allows more CA to be impregnated into paper, and thus more fibre-CA bonds formed (Fig. 2). The additional bonds improve the bonding strength among the fibre of the paper and consequently increase the burst index. The same effect is also observed when *C* is increased. The fact that the graph is curved across factor *T* demonstrated that *T* gives the best effect relative to burst index, in the region of high *D* and high *C*, it can be observed that the increase of *T* until 140 °C increased the burst index significantly. Nevertheless, it starts to give an inverse result when *T* is higher than 140 °C. It is proposed that *T* above 140 °C has an adverse effect on the additional fibre-CA bonding.





Effect on rate of surface wettability

Commercial Braille paper has very high water absorbability, and therefore, the embossed Braille dots will be blemished when the paper gets wet. Low water absorbability or rate of surface wettability is a property aimed to be added to the Braille paper in this study so that the Braille dots can still be sustained even if it is splashed with water. Table 1 shows that the wettability values of CA-impregnated paper were in the range of 0.010 to 0.10°/s, which is much lower than that of commercial Braille paper (0.418°/s) and also the non-impregnated handsheet (Table 4). This indicated that impregnation with CA decreases the rate of surface wettability effectively. However, the wettability property is influenced by the variables associated with the impregnation process employed.

Based on the statistical analysis, D and T were found to give significant effects relative to the rate of surface wetting. C alone had an insignificant effect on the rate of surface wetting, but the interaction effects of TC and DC were significant. It is very interesting to see that when C is low, the rate of surface wettability increases from the region of low T and high D toward the region of high T and low D. However, the graph totally reverses when C is high, as shown in Fig. 5. This revealed that the water resistance was high when the conditions of low C, low T, and high D or high C, high T and low Dwere employed. In comparison to Fig. 2, it was found that the papers impregnated by using the two conditions with high water resistance did not possess higher percentage of CA. This indicated that the amount of CA in the paper did not influence the rate of surface wettability. Since the results were out of expectation, it is still hard to explain this phenomenon at this stage.



Fig. 5. 3D response surface plot of rate of surface wettability as a function of dipping time and temperature with a fixed percentage of concentration of cellulose acetate at 1.8, 2.5, and 3.2 percent

Optimization

Based on the statistical experimentation, it was shown that recycled paper impregnated with CA made it possible to enhance paper properties. In order to obtain the optimum impregnation condition, an optimization process is carried out. The desired paper properties and the calculated optimum conditions suggested by Design-Expert® are shown in Table 5 and Table 6, respectively. A condition (the highlighted one) among the calculated conditions is selected, and experimentation is carried out in order to verify the reliability of the estimation models. The values of the predicted and actual experimental results are shown in Table 7. The experimental results of smoothness, burst index, and rate of surface wettability were in the range of the Predicted Interval (PI) results. However, the result of density was not in the range of PI, which is only 2 percent lower than PI. This is possibly due to the fact that the handsheet made from secondary fiber had been kept for more than six months.

Name	Goal	Lower Limit	Upper Limit					
Density	is in the range	0.55	0.6282					
Smoothness/Roughness	is in the range	214	400					
Burst Index	is in the range	2.2	3.61					
Rate of Surface Wetting	is in the range	0	0.55					

Table 5. Desired Goal of Paper Properties

Tab	Table 0. Calculated Optimum Conditions Trovided by Expert Designe									
No.	Temp.	Dipping Time	Conc. Of CA	Density	Smoothness/ Roughness	Burst Index	Rate of Surface Wettability	Desirability		
1	163.76	2.82	2.70	0.589989	397.051	2.2	0.050	1		
2	123.80	8.07	1.88	0.556533	396.816	2.3	0.044	1		
3	110.13	8.89	2.06	0.551924	367.438	2.4	0.044	1		
4	105.93	9.49	2.20	0.550855	372.503	2.5	0.048	1		
5	106.43	10.03	2.22	0.550637	383.726	2.5	0.046	1		
6	163.82	2.60	2.79	0.588875	398.744	2.2	0.046	1		
7	116.08	9.52	2.13	0.556385	394.694	2.5	0.044	1		
8	114.25	9.19	1.92	0.550236	383.838	2.3	0.036	1		
9	115.91	9.08	2.05	0.555186	386.136	2.4	0.043	1		
10	163.98	2.71	2.71	0.590033	396.159	2.2	0.049	1		

Table 6. Calculated Optimum Conditions Provided by Expert Design®

Table 7. Comparison Between Predicted and Actual Experimental Values

	- Exporimont			
Responses	Prediction	95%	95%	Experiment
	Value	PI low	PI high	
Smoothness / Roughness	397	207	585	475
Burst Index	2.2	1.5	2.9	2.84
Rate of Surface Wettability	0.05	0.007	0.093	0.012
Density	0.5899	0.5584	0.6212	0.5450

CONCLUSIONS

The statistical assessment of response surface methodology revealed that among the three independent variables of the impregnation condition, the dipping time was the most influential variable relative to the four responses (density, smoothness, burst index, and rate of wettability), followed by the pressing temperature and the concentration of cellulose acetate. Based on the estimation models obtained, the optimum impregnation condition involved dipping the handsheet in a 2.7 percent cellulose acetate solution for 2.8 minutes and then pressing at 163 °C. The resultant Braille papers made from secondary fibre and impregnated with cellulose acetate exhibited better properties in terms of rate of surface wetting and burst index than those of commercial Braille paper. Although the CA impregnated handsheets still had a lower density, the stiffness of Braille dots was better than the non-impregnated hand sheet. As the grammage of impregnated hand sheet is lower than the commercial Braille paper, it helps to lessen the burden of users as the total weight of Braille books become lighter. The density and smoothness of the produced Braille paper still have room of improvement in order to achieve the 0.75 g/cm² and 131 mL/min of commercial Braille paper.

ACKNOWLEDGEMENTS

The authors would like to acknowledge Professor Dr. Abdul Khalil Swakataly for his advice on the determination of the surface wettability of paper. The precious advice and comments from St Nicholas Home Penang, SMK Alma, SMK HAJI Zainal Abidin, Mr. Ooi Hock Tiam (School of Humanities, USM), Dr. Mellisa Ng Lee Yen Abdullah (School of Educational Studies, USM) along the research are greatly appreciated. Financial support from Universiti Sains Malaysia (Intensive grant and RU fellowship (1001-PTEKIND/821067 and 1001/PTEKIND/814164) is gratefully acknowledged.

REFERENCES CITED

- Baker, W. O., Fuller, C. S., and Pape, N. R. (1942). "Effect of heat, solvent and hydrogen bonding agents on the crystallinity of cellulose esters," *J. Am. Chem. Soc.* 64(4), 776-782.
- Casey, J. P. (1981). *Pulp and Paper Chemistry and Chemical Technology* 3rd Edition, Wiley-Interscience New York.
- Cox, S. T., Mitchell, M. G., Kennedy, P. J., Massie, Jr., G. F., and Williams, F. W. (1997). "High pressure laminates made with paper containing cellulose acetate," U.S. Patent 5,928,777, Jun.12, 1997.
- Eastman (2006). "Product data sheet eastman cellulose acetate," (CA-398-3): http://ws.eastman.com/ProductCatalogApps/PageControllers/ProdDatasheet_PC.aspx ?product=71001217
- Filho, G. R., Monteiro, D. S., da Silva, M. C., Nascimento de Assunção, R. M., Alves Cerqueira, D., Barud, H. S., Ribeiro, S. J. L., and Messadeq, Y. (2008). "Synthesis

and characterization of cellulose acetate produced from recycled newspaper," *Carbohydrate Polymers* 73(1), 74-82.

- Frederick, T. J., Mitchell, M. G., and Partin, L. R. (1997). "Paper made with cellulose fibres having an inner core of cellulose acetate," U.S. Patent 5573640, Feb. 13,1997
- Ibrahem, A. A., Nada, A. M. A., El-Meadawy, S. A., and Yousef, M. A. (1983). "Improvement of paper properties by impregnation in cellulose derivative solution," *Acta Polymerica* 34(3), 178-180.
- Keith, C. H. (1977). "Cellulose ester fibrillar structure," U.S Patent 4,047,862, Sept.13, 1977.
- Law, R. C. (2004). "Applications of cellulose acetate. 1: Cellulose acetate in textile application," *Die Makromolekulare Chemie. Macromolecular Symposia*, Wiley Inter-Science, Weinheim.
- Litzinger, P. C. (1977). "Production of discrete cellulose acetate fibers by emulsion flashing," US patent no. 4040856.
- Loo, M. M. L., Hashim, R., and Leh, C. P. (2012). "Recycling of valueless paper dust to a low grade cellulose acetate: Effect of pretreatments on acetylation," *BioResources* 7(1), 1068-1083.
- Mitchell, M. G., Kirk, W. H., Crosswhite, C. K., and Kennedy, P. J. (2000). "Multiply paper comprising a mixture of cellulose fiber and cellulose ester fibers having imparted softening properties and a method of making the same," *US Patent* 6,010,595.
- Rex, E. H., Loenig, A. J., Wormsley, D. P., and Baker, R. L. (1995). Foundations of Braille Literacy: A History of Literacy for People Who are Blind, AFB Press, New York, pp. 15-33.
- Rustemeyer, P. (2004). "History of CA and evolution of the markets," *Macromol. Symp.* 208, 1-6.
- SCAN-P 84:02 "Surface roughness and surface compressibility. Bendtsen method with pressure correction".
- Sjöholm, E. (2004). "Size exclusion chromatography of cellulose and cellulose derivatives," *Handbook of Size Exclusion Chromatography and Related Techniques*, Chapter 12, Marcel Dekker, NY, USA.
- Soo, M. C., Loo, M. M. L., Wan Rosli, W. D., and Leh, C. P. (2010). "The effect of low grade cellulose acetate coating variables in the production of braille paper from secondary fibre studied by fractional factorial design," International Conference of Environmental Research and Technology, 332-338.
- TAPPI Useful Method T403 "Bursting strenth of paper," Technical Association of The Pulp and Paper Industry, Atlanta.
- TAPPI Useful Method T411 "Thickness (calliper) of paper, paperboard, and combined board," Technical Association of The Pulp and Paper Industry, Atlanta.
- TAPPI Useful Method T458 "Surface wettability of paper (angle of contact method)," Technical Association of The Pulp and Paper Industry, Atlanta.

Article submitted: November 10, 2011, Peer review completed: July 2, 2012; Revised version received: July 24, 2012; Accepted: August 4, 2012; Published: September 13, 2012.