The Optimization of the Water Footprint and Strength Properties of Handsheets by the Extreme Vertices Mixture Design

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Water footprint (WF) is widely used as a life cycle assessment (LCA) tool to assess the environmental impacts of water usage associated with forestry-based production. The calculations of WF are significantly influenced by the raw materials and the process. Some information is available on WF in the papermaking industry. However, there has been little consideration of the correlation between the WF and the properties of paper. Technically, the WF and the properties of paper are impacted by the raw materials. Generally, the ideal formula of raw materials used to make paper could decrease the WF while maintaining the properties of the paper. In the current study, the extreme vertices mixture design was used to optimize the WF and properties of the handsheet by the raw materials. The new model indicated that the WF of the handsheet was decreased significantly while the properties was maintained through the adjustment of the raw materials.

Keywords: Water footprint; Extreme vertices mixture design; Strength properties; Handsheet

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INTRODUCTION

As concerns for water scarcity and pollution increase, interest in a new means of water resource management such as the water footprint, has grown rapidly (Hoekstra 2009; 2030 Water Resources Group 2009). A water footprint (WF), *e.g.* an operation water footprint or a supply chain water footprint, maps the impact of human activities on fresh water resources and the environment (Vince and Koehler 2010; Wessman 2011; Hoekstra and Mekonnen 2012). According to Hoekstra's definition, a water footprint includes the green water footprint, blue water footprint, and gray water footprint (Chapagain and Hoekstra 2011; Hoekstra *et al*. 2012). The green water footprint refers to the amount of rainwater consumed during the agricultural or forestry-based production process, *i.e.*, the total rainwater evapotranspiration from soil and plantation plus the water incorporated into vegetation. The blue water footprint is an indicator of consumptive use of fresh surface and groundwater. The consumptive use means evaporated, incorporated into product, or not immediately returned within the same catchment or aquifer. The grey water footprint is an indicator of the degree of freshwater pollution defined as the amount of freshwater that is required to assimilate the load of pollutants based on natural background concentrations and existing ambient water quality standards. Among them, the green water footprint contributes more to the water footprint in processes where agricultural or forestry-based products are used as raw materials (Mekonnen and Hoekstra 2010, 2011a). Currently, many companies have realized that the usage of water in the supply chain (defined as the indirect water footprint) is much greater than that in the operation process (defined as the direct water footprint) in industries such as pulp and paper manufacturing, bio-energy production, and textiles. To minimize the water footprint of the whole process, it is just as important to decrease the water footprint of the supply chain and the associated risks as it is to manage the operation water usage (Hastings and Pegram 2012; Postle *et al*. 2012).

Most of the attention in traditional water resource research and management has focused on the blue water footprint, which technically underestimates the importance of green water as a contributor to the water footprint. Technically, blue water and green water can be transformed into each other through the earth's hydrologic cycle. Green water can be used to replace part of blue water in parts of agriculture and wood production processes. Hence, a complete map of the water footprint can be obtained by taking both the blue water footprint and green water footprint into consideration. It is well known that quite large amounts of fresh water from rivers, lakes, and aquifers are consumed in the pulp and paper-making industries. At the same time, trees that are used as the starting material in the pulp and paper industries also consume large amounts of fresh water during their growth (Stora Enso 2011; Rep 2011). Therefore, the water footprint of the entire process, including the supply chain water including raw materials and chemicals and the operation water, can help to make sustainable water management more effective in papermaking industry (Shen and Qian 2012; Manzardo *et al*. 2014) .

The extreme vertices mixture design has been used to elucidate the correlation between investigated variables and results (Vagas *et al*. 2008; de Oliveira *et al*. 2011). This method is capable of optimizing several variables simultaneously and obtaining the best response, especially in the examination of food, beverage, steel, and chemical manufacturing (Unger *et al*. 2013). Water footprint has been applied as a life cycle assessment tool to evaluate the environmental impacts of water usage in paper industry (Berger and Finkbeiner 2010; Kounina *et al*. 2013). In this study, the extreme vertices mixture design was applied to elucidate the effect of raw materials WF on the final product's WF by the different formulations of a paper handsheet. The correlation between raw materials and water footprint of handsheets produced in our lab were analyzed using the Minitab software version 16.

EXPERIMENTAL

Materials

All chemicals and raw materials were obtained from Paper Machine #2 (PM#2) in Gold East Paper Company (Zhenjiang, China). Softwood bleached kraft pulp (NBKP, "needles"), hardwood bleached kraft pulp (LBKP, "leaves"), alkaline peroxide mechanical pulp (APMP), ground calcium carbonate (GCC), and cationic cassava starch were used to make the handsheets. The sum of the five raw materials was set to 100%. The NBKP, LBKP, and APMP used in this study were obtained from Canada, Indonesia, and China, respectively. The NBKP and LBKP were refined to a freeness of 370 to 390 mL CSF and 392 to 402 mL CSF, respectively. The solids contents of the cationic starch solution and the pre-dispersed ground calcium carbonate (GCC) slurry were 4.00% (wt.) and 20.00% (wt.), respectively.

Table 1. Variables and Levels of Extreme Vertices Mixture Design for Handsheet Preparation

Components	Lower (%)	Upper (%)
NBKP	20.00	50.00
LBKP	30.00	60.00
APMP	10.00	40.00
GCC	10.00	40.00
Cationic Starch	0.20	2.00

Table 2. Dosages of Chemical Additives

Chemicals	Dosage
Wet Strength Agent (%)	0.5
Dry Strength Agent (%)	1.0
Retention Aids (ppm)	300.0
Bentonite (ppm)	4500.0
Polyacrylamide (ppm)	300.0

Note: Values are based on dry fiber mass

Methods

Water Footprint of Components

Renewable green water is used by rain-fed forests in the root zone (Samuli *et al*. 2013). However, pulp mills usually use blue water, of which the amount consumed throughout the pulping process is much less than the volume of water needed for forestry growth. Thus, it is reasonable to consider only the green water footprint of the fiber stock while neglecting the blue and gray water footprints of the pulping process in this study. The green water footprint of cationic cassava starch evaluated by Mekonnen and Hoekstra 2011b) was applied directly in this investigation. The blue water footprint of GCC was calculated using the following equation,

$$\mathrm{WF_{GCC,Blue}} = \frac{\mathrm{WW_{fresh}} - \mathrm{E}}{\mathrm{P}} \tag{1}$$

where $\mathrm{WF_{GCC,Blue}}$ is the blue water footprint of GCC ($m^3\cdot\cdot t^{-1}$), $\mathrm{WW_{fresh}}$ is the monthly fresh water used in a GCC plant ($m^3\cdot\cdot month^{-1}$), E refers to the monthly effluent in a GCC plant ($m^3\cdot\cdot month^{-1}$), and P refers to the production of GCC ($t\cdot\cdot month^{-1}$).

Technically, the WF of paper includes the water footprints of both raw materials and water consumed during the entire process. However, only the water footprint of raw materials used to make the handsheets (indirect water footprint) was studied in this investigation. Thus, the WF of a handsheet is the sum of the green water footprints of fiber and cationic starch and the blue water footprint of GCC. The green water footprint of raw materials arising from the evapotranspiration of forests (Van Oel and Hoekstra 2010) represents the largest component of water resources consumed in the pulp and paper industry. The water footprint of forestry is estimated using the following equation (Van Oel and Hoekstra 2012),

$$\mathrm{WF_{foresty}} = \left[\frac{\mathrm{WWU_{green}} + (Y_{\mathrm{wood}} \times f_{\mathrm{water}})}{Y_{\mathrm{wood}}}\right] \times f_{\mathrm{pulp}} \times f_{\mathrm{value}} \times \left(1 - f_{\mathrm{recycling}}\right) \quad (2)$$

where $\mathrm{WWU_{green}}$ is the green water component of wood water use in a forest/woodland (m^3/ha/year), Y_{wood} is the wood yield from a forest or woodland (m^3/ha/year), f_{water} is the volumetric fraction of water in freshly harvested wood (m^3/m^3), f_{pulp} is the wood-to-pulp conversion factor, f_{value} is the fraction of the total value of the forest that is associated with paper production, and $f_{\mathrm{recycling}}$ is the fraction of pulp derived from recycled paper. The value of $\mathrm{WWU_{green}}$ is calculated by accumulation of daily evapotranspiration ($\mathrm{ET_{green}}$, mm/day) over the complete growing period of forests and is calculated using the following equation (Hoekstra and Chapagain 2011),

$$\mathrm{WWU_{green}} = 10 \times \sum_{\mathrm{d=1}}^{\mathrm{lgp}} \mathrm{ET_{green}} \quad (3)$$

where $\mathrm{ET_{green}}$ is green water evapotranspiration and 10 is the factor converting water depth in millimeters into water volume *per* land surface in m^3/ha. The summation is done over the period from the day of planting (day = 1) to the day of harvest (lgp stands for length of growing period in days).

Eventually the WF of handsheets can be calculated as the sum of all starting materials,

$$\mathrm{WF_{handsheets}} = \sum_{\mathrm{i=1}}^{5} \mathrm{WF_i} \times C_{\mathrm{i}} \quad (4)$$

where $\mathrm{WF_i}$ is the water footprint of the i^{th} raw material, and C_{i} is the percentage of the i^{th} raw material.

Design of experiments

An experimental design resulting in 17 handsheets with various contents of components at a basis weight of 100 $g \cdot m^{-2}$ was achieved by MINITAB version 16 of Minitab, Inc., USA using an extreme vertices mixture design. Five variables, *e.g.*, NBKP, LBKP, APMP, GCC, and cationic starch were used to elucidate their impacts on strength properties and WF of handsheet. Variables, representing low and high levels of extreme vertices mixture design for handsheet preparation are shown in Table 1. The detailed dosages of other chemical additives and the values of component used to prepare handsheets are shown in Tables 2 and 3, respectively.

A mixture regression fitting method was applied to investigate how these five variables affected strength properties of prepared handsheets, and a quadratic model was employed from six standard models (linear, quadratic, special cubic, full cubic, special quartic, full quartic) supplied by statistical software Minitab 16. Both strength properties and WF of a handsheet depends on the relative proportions of the components that are changed at the interval of low level and high level, but the sum of all investigated variables proportion is kept at 100%. The desired confidence level is 95% (95% CI) as the default value in Minitab statistical software, which means that the significance level is set as 0.05 in the multivariate F-test. The effects of five components on strength properties and WF were analyzed by response trace plot, contour plot, and overlaid contour plot supplied by statistical software Minitab 16.

Strength properties of prepared handsheets including tensile, tear, and burst strengths were examined by the standard methods TAPPI T494 om-01 (2001), TAPPI T414 om-04 (2004), and TAPPI T403 om-02 (2002).

Table 3. Component Formula of Handsheet Preparation

Sample No.	NBKP (%)	LBKP (%)	APMP (%)	GCC (%)	Cationic Starch (%)
1	23.6125	33.6125	28.5125	13.6125	0.6500
2	49.8000	30.0000	10.0000	10.0000	0.2000
3	20.0000	30.0000	10.0000	38.0000	2.0000
4	20.0000	30.0000	10.0000	39.8000	0.2000
5	23.6125	33.6125	27.6125	13.6125	1.5500
6	20.0000	58.0000	10.0000	10.0000	2.0000
7	23.6125	48.5125	13.6125	13.6125	0.6500
8	48.0000	30.0000	10.0000	10.0000	2.0000
9	23.6125	47.6125	13.6125	13.6125	1.5500
10	27.2250	37.2250	17.2250	17.2250	1.1000
11	38.5125	33.6125	13.6125	13.6125	0.6500
12	20.0000	30.0000	39.8000	10.0000	0.2000
13	23.6125	33.6125	13.6125	28.5125	0.6500
14	37.6125	33.6125	13.6125	13.6125	1.5500
15	20.0000	30.0000	38.0000	10.0000	2.0000
16	20.0000	59.8000	10.0000	10.0000	0.2000
17	23.6125	33.6125	13.6125	27.6125	1.5500

RESULTS AND DISCUSSION

Water Footprint of Raw Materials

The WF of fiber was calculated according to Eq. 2. The values of ET_{green}, Y_{wood}, f_{water}, and f_{pulp} were adopted from previous studies (Van Oel and Hoekstra 2010; 2012; Hoekstra and Chapagain 2011). Because the handsheet is the only product in this investigation, the value of f_{value} was set to one. The value of $f_{recycling}$ was zero because little recycled pulp or paper was used. Other WFs of raw materials applied in this study are shown in Table 4.

Table 4. Water Footprint of Raw Materials

Raw Materials	WF ($m^3 \cdot t^{-1}$)
LBKP	3311.6
NBKP	2695.8
APMP	1852.3
Cationic Starch	2254.0
GCC	0.2

Properties and WF of Handsheets

The strength properties and WF of the prepared handsheets are shown in Table 5. The strength properties of all prepared handsheets were characterized according to the standard testing methods TAPPI T494om-01 (2001), TAPPI T414om-04 (2004), and TAPPI T403om-02 (2002). The WF of each handsheet prepared with various components was calculated using the previous equations.

Table 5. Handsheet Strength Properties and WF

Sample No.	Tensile Index ($N \cdot m \cdot g^{-1}$)	Tear Index ($mN \cdot m^2 \cdot g^{-1}$)	Burst Index ($kPa \cdot m^2 \cdot g^{-1}$)	WF Volume ($m^3 \cdot t^{-1}$)
1	42.42	6.03	2.43	2489.4
2	58.08	7.26	3.81	2830.3
3	34.71	5.24	1.92	2479.3
4	26.12	5.53	1.35	2519.4
5	47.04	6.87	2.88	2473.0
6	62.74	7.25	3.96	2601.4
7	47.75	7.75	2.81	2613.4
8	67.14	8.02	4.38	2771.5
9	54.76	7.36	3.43	2589.5
10	49.27	7.38	3.12	2577.6
11	56.50	8.35	3.43	2704.0
12	38.99	7.84	2.05	2401.2
13	38.63	7.00	2.33	2548.5
14	58.48	8.09	3.92	2674.6
15	46.65	6.87	2.90	2368.4
16	49.06	9.19	3.02	2649.3
17	41.23	6.56	2.32	2528.5

Impacts of Components on Strength Properties

The response trace plot method was applied to statistically analyze the effect of raw materials on the strength properties and WF of handsheets in this study. Theoretically, a response trace plot can show the effect of each component on the corresponding response. Several response traces, which are a series of predictions from the fitted model, are plotted along a component direction. The trace curves indicate the effect of changing the corresponding component along an imaginary line (direction). Thus, each component of the handsheet formula has a corresponding trace. The points along a trace direction of a component are connected, thereby producing as many curves as there are components in the mixture. Response trace plots are especially useful when there are more than three components in the mixture and the complete response surface can be visualized on a contour or surface plot. The impact of components on strength properties was evaluated by a response trace plot. Each formulation component in the mixture had a trace that represented its variation along each axis (de Oliveira 2011; Kleemann 2012). The response trace plots for tensile, tear, and burst indices are exhibited in Figs. 1a, 1b, and 1c, respectively, which indicate the correlations between components and strength properties. The results suggested that GCC content had a strong negative influence on the strength properties; however, NBKP and LBKP had a positive impact. Meanwhile, there was no simple correlation identified between APMP/cationic starch and strength properties.

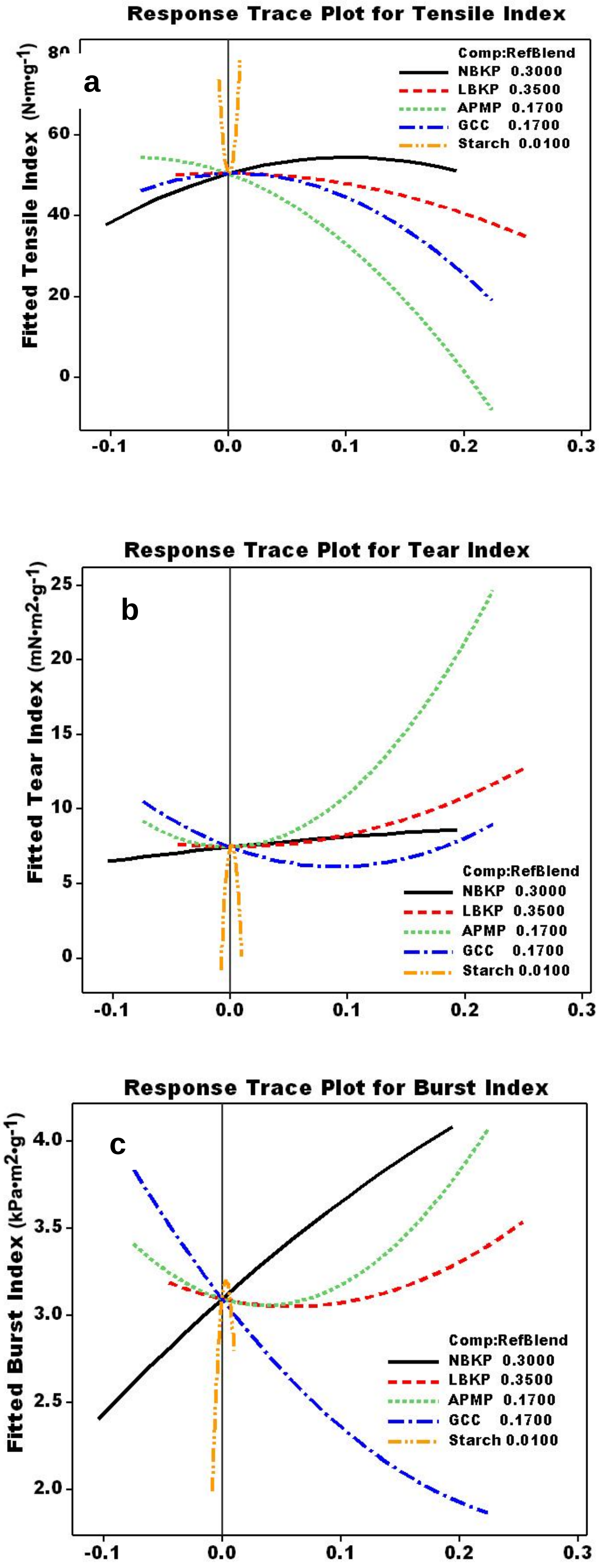

Fig. 1. Response trace plot showing the correlation between the raw material and strength properties: (a) tensile index; (b) tear index; and (c) burst index

Theoretically, it is expected that the application of starch can improve the strength properties of paper because of the enhancement of hydrogen bonding between fibers (Hubbe 2006). However, the strength properties of paper decreased as more APMP was involved because its fiber length is relatively short. Technically, the strength properties of paper are significantly impacted by hydrogen bonding and fiber length (Ekhtera *et al*. 2008; Brännvall 2009).

Impacts of Components on Water Footprint

The impact of components on the water footprint is demonstrated in Fig. 2. In the response trace plot, all components were interpreted relative to the reference blend, and its value is shown in the legend. With increasing amounts of APMP, cationic starch, and GCC in the handsheets, the WF decreased, and yet it increased when more NBKP or LBKP was used.

These results can be explained by the range of the component (upper bound - lower bound), the direction, and steepness of its response trace. The LBKP had a stronger positive effect on the WF of the prepared handsheets compared to NBKP, which is confirmed by its sharper slope. The APMP showed a negative effect, followed by GCC. The impacts of raw fiber materials on the WF of the handsheets were analyzed using the contour plots method (Fig. 3). For instance, the WF changed between 2750 and 2500 $m^3 \cdot t^{-1}$ according to the content of APMP and LBKP, with the content of NBKP kept constant. However, the WF only changed between 2750 and 2650 $m^3 \cdot t^{-1}$ according to the content of NBKP and LBKP when the content of APMP remained constant.

Combining the strength properties analysis and the WF results, it is proposed that raw materials with lower WFs should be used to obtain the paper with desired strength properties and less water footprint. The obvious scheme was to use materials having lower water footprint as much as possible, and contour plots provided more accurate analysis and optimization.

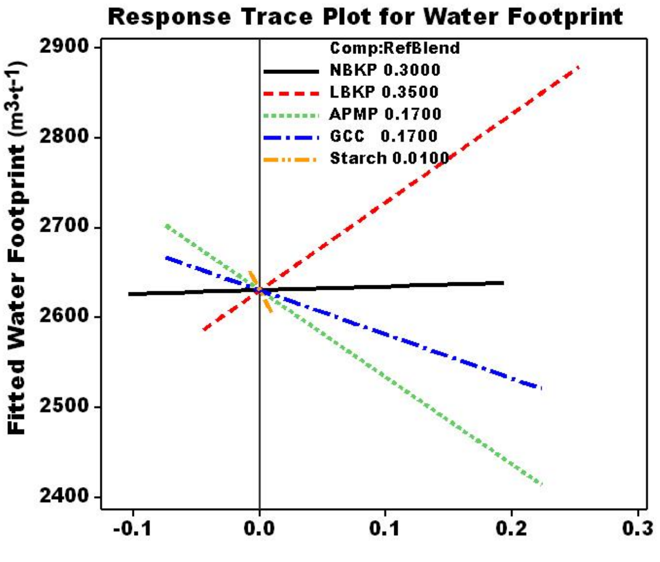

Fig. 2. Response trace plot showing the correlation between raw material and WF

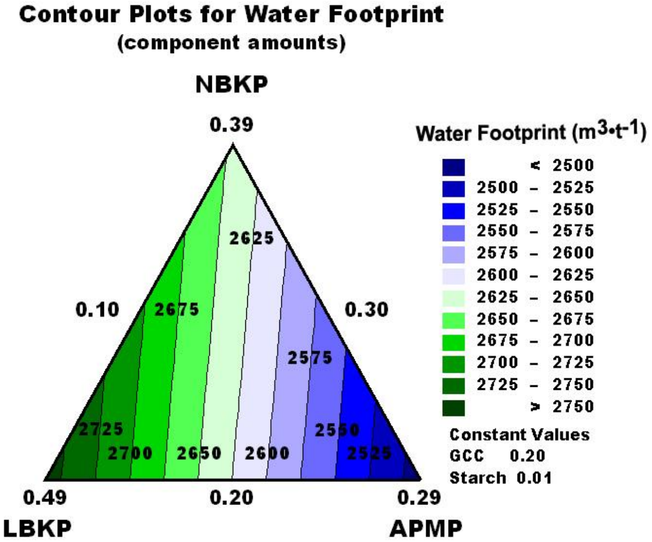

Fig. 3. Contour plot showing the correlation between raw material and WF

Water footprint is a comprehensive indicator of freshwater resource appropriation that includes the direct water usage and the indirect water usage, which differs from the traditional measure of water use in a paper mill (Nalco 2011). Reduction of fresh water usage and improvement of strength properties of the paper product have been studied for many years in the pulp and paper industry (Mänttäri *et al*. 2002; Pizzichini *et al*. 2005). It had been reported that the raw materials significantly affect the water footprint and the strength properties of the final products (Van Oel and Hoekstra 2012; Manzardo *et al.* 2014). Based on the reported results it was attempted to maintain the strength properties while minimizing the WF of handsheets at the same time using an extreme vertices mixture design in this study. In Fig. 4, Circle 1 represents all potential component formulations of handsheet formation.

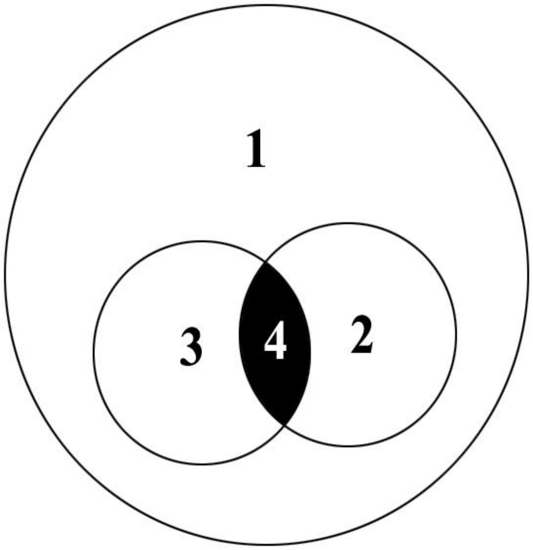

Fig. 4. Schematic process of optimization of strength properties and WF

Circle 2 represents some formulations of handsheets that have the desired strength properties; Circle 3 represents some formulations of handsheets that have the desired water footprint; and intersection 4 is the specific formulation required to make handsheets with optimized strength properties and water footprint.

It is of little value to enhance the strength properties of handsheets by increasing NBKP and LBKP or decreasing GCC content because these actions lead to a high WF of the final product. Therefore, it can be realized to minimize the water footprint of the handsheet while maintaining the desired strength properties of handsheet through decreasing the content of fibers in the formula. In the overlaid contour plot (Fig. 5), the borders of the contour lines for each response are defined, and the blank area is the specific formulation of components, including NBKP, LBKP, and ATMP, to achieve handsheets with the desired strength properties. The optimization of the strength properties and WF of handsheets is presented in Fig. 6. The composite desirability of the four response variables (tensile strength, tear strength, burst strength, and WF) was 0.799. The individual desirabilities for the tensile strength, tear strength, burst strength, and WF of the handsheets were 0.598, 0.992, 1.000, and 0.687, respectively. As a result, the optimal proportions of the five components used to make handsheets are as follows: 48% NBKP, 30% LBKP, 10% APMP, 10% GCC, and 2% cationic starch. Consequently, the optimal strength properties and WF of the handsheet were predicted to be 66.92 N·m·g^{-1} (tensile strength), 7.96 mN·m^{2}·g^{-1} (tear strength), 4.39 kPa·m^{2}·g^{-1} (burst strength), and 2663 m^{3} ·t^{-1} (WF).

A test was conducted using the optimal formulation to verify if the strength properties were consistent with the predicted results (Table 6). As expected, all test results fell into the 95% confidence level (95% CI) of predicted responses indicated that the extreme vertices mixture design could be used to set up the component formula for handsheets to minimize its WF and maintain its optimal strength properties.

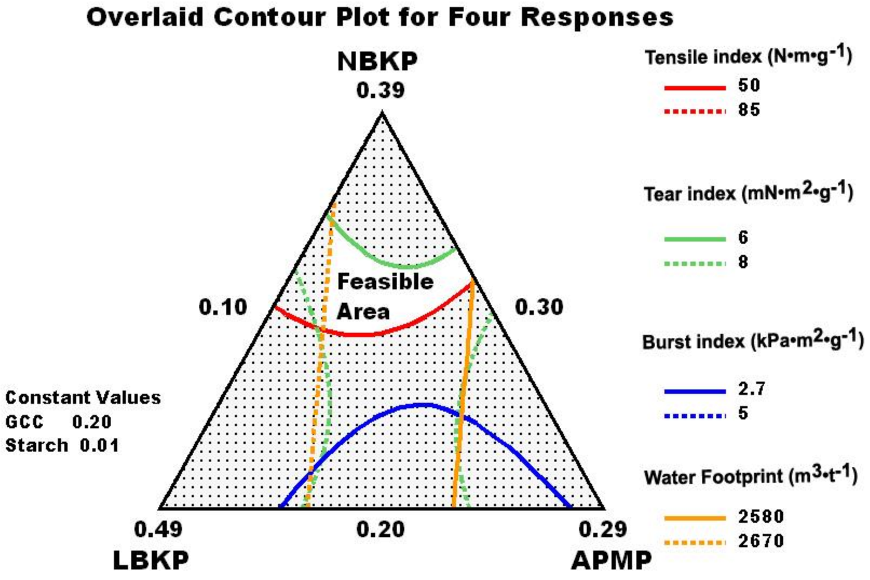

Fig. 5. Overlaid contour plots showing tensile strength, tear strength, burst strength, and WF

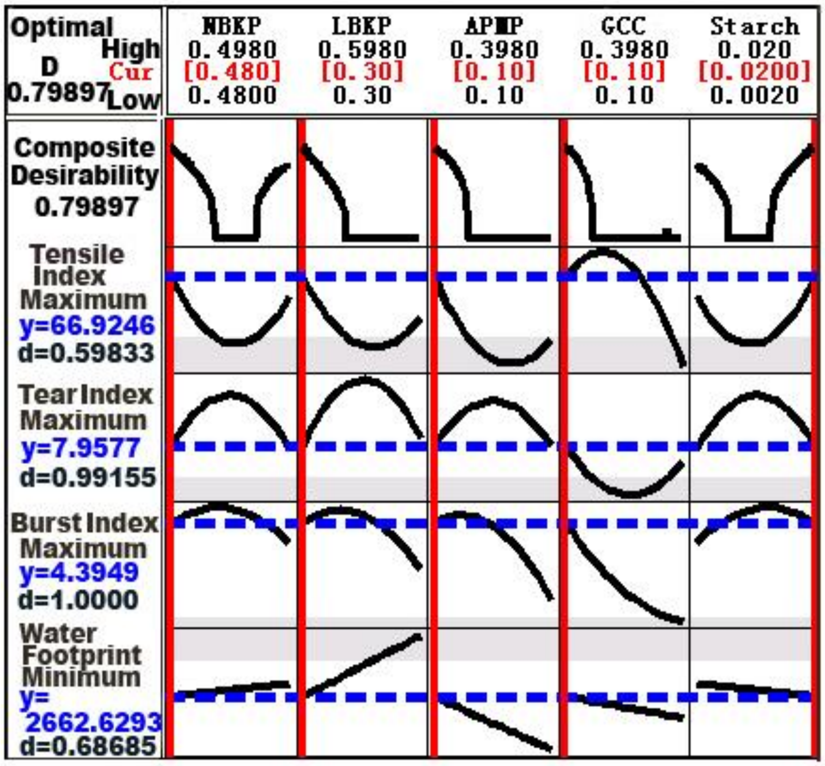

Fig. 6. Optimization plot showing tensile strength, tear strength, burst strength, and WF

Table 6. Predicted Response and Mean Verification of Strength Properties

Properties	95% CI	Mean Verification Results
Tensile Index (N·m·g^{-1})	(54.36, 83.08)	57.54
Tear Index (mN·m^2·g^{-1})	(2.85, 13.14)	8.59
Burst Index (kPa·m^2·g^{-1})	(1.76, 6.22)	2.76

CONCLUSIONS

1. An empirical case was investigated to minimize the water footprint of handsheets while maintaining the strength properties of the paper. As a result, a mathematical model is presented in the current study that could be used to optimize the WF and strength properties of handsheets.
2. The components and content of raw materials used to make the handsheets influenced not only the strength properties but also the water footprint. Thus, the handsheet was produced by the raw materials that could minimize the WF of the handsheet while maintaining the optimal strength properties of the handsheet. According to our model, the optimized component formula used to make a handsheet with the desired strength

properties and WF was 48% NBKP, 30% LBKP, 10% APMP, 10% GCC, and 2% cationic starch.

3. The verification tests were conducted to confirm the optimized component formulation. The predicted response demonstrated a confidence level of 95% for the statistical analysis. Technically, the results suggest that the extreme vertices mixture design is an effective method to evaluate the effect of raw materials on the water footprint of handsheets. It is noteworthy that the prediction of WF could be varied if one or more factors was changed. Thus, this study only provides the state of art statistics model for water footprint analysis of paper in the papermaking industry.

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