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EVALUATION OF COATED PAPER QUALITY USING A PLACKETT-BURMAN STATISTICAL DESIGN

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Paper is pigment-coated to produce a more uniform and more receptive surface for printing ink, thus assuring better graphic reproduction. A large number of variables, involved in coating processes, interact with each other. To perform coating experiments on indigenous base papers in a most systematic way, with many variables, statistically designed experiments – based on a Plackett-Burman design – have been carried out to examine the effects of different process variables. The variables considered were the following: dosages of china clay, talc, natural and precipitated calcium carbonate, binder, as well as total solids of coating color and thickener dosage (water retention chemicals). Based on such experiments, the effects of the above process variables on different properties of coated paper were identified.

Keywords: coating, statistical design, precipitated calcium carbonate, brightness, talc, gloss

INTRODUCTION

Coating of paper represents a layered construction and a multi-variable system, which makes the quantitative estimations on the effect of each parameter of the process very difficult. The relative effect of so many variables on the properties of paper requires systematic investigation, to obtain quantitative estimates of the effect of each parameter upon the process, which is really a Herculean task. The complexity of the situation arises from the interaction of variables, which cannot be separated from their main effect. In such an intricate situation, statistical modeling can be of enormous use. It has already been proved that statistical modeling can provide a means for reducing the enormous number of experiments normally required in traditional testing. In traditional experiments, only one variable can be changed at a time, on keeping the others constant, which may lead to erroneous conclusions. This situation can be avoided by various designs of the experimental methodology, which not only avoid laborious experimentation, but also provide a better quantification of parameter relationship. As a matter of fact, different coating color formulations have been made[1-3] by different statistical experimental design techniques, such as full factorial design, the methods of McLean Anderson, Scheffe, Draper Lawrance, mixture design, etc. Out of all statistical design techniques, the most important is that of Plackett-Burman (P-B), generally preferred in the evaluation of the effect of process variables.[4]

Plackett-Burman experimental design

The P-B design[5,6] is a two-level D-optimal saturated orthogonal design (fractional factorial) constructed on the basis of fractional replicates of a full factorial design. Further on, it is based on balanced incomplete blocks and on the use of N experiments (N number of runs) to study the process variables (k = N – 1), where N is a multiple of 4. For obtaining an orthogonal design matrix, the following conditions are necessary and sufficient:

1. The number of times each factor is adjusted to each of its levels must be the same;

2. The number of times every two factors, each at any one of its levels are encountered, must be the same;

3. The number of observations must be divisible by the square of the number of levels, defined as:

$$N = nl^2 \quad (1)$$

where n is an integer. When the above-stated conditions are available, the construction of an orthogonal matrix (experimental design) requires combinatorial operations only.

If $N = nl^2$, then the number of factors for which one can compute their effects, a given n, is:

$$k = (nl^2 - 1)/(l - 1) \quad (2)$$

or an integer part of the value given by equation (2).

If the number of levels for all factors is 2, then an optimum design is obtained by constructing an N x N orthogonal matrix comprising +1's and -1's, where N is a multiple of 4, *i.e.* N = 4n. The maximum number of factors that can be included in the design is k = N – 1.

Again, if the difference between the effects of each primary variable is not sufficient to permit confident identification of the important factors by inspection, an alternative is to construct normal or half-normal probability plots of the effect. In these plots, the ordinate represents the cumulative percentage probability, P, with m data points taking values for constant spacing:

$$P_i = 100(I - ½)/m, \text{ where } I = 1, 2…m \quad (3)$$

over the cumulative distribution. The ordinate scale is adjusted to linearize the ideal sigmoid curve, so that if all effects were normally distributed, they would be plotted as a straight line. Thus, major deviations from this straight line indicate that these effects are not easily explained as random occurrences.

An incomplete factorial design, which measures only the main effects (no interaction), is actually the P-B design, in which the number of experiments is greatly reduced, but useful data are generated. Because these designs cannot be represented as cubes, they are sometimes called non-geometric designs. Each variable is at one of two levels with high (+) and low (-). These levels were selected far enough apart, to expect a significant response in coating, but not so remote from normal conditions. (*i.e.* if the levels are set too close together or too far apart, they may give a non-significant response difference, whereas the factor effect may be significant).

A major disadvantage of the factorial design is that an increase in the number of factors is accompanied by a dramatic rise in the number of experiments. As an example, the five-factor two-level unreplicated designs require $2^5 = 32$ experiments, and the four-factor three-level design requires $3^4 = 81$ experiments. This difficulty can be overcome by reducing the number of experiments without a substantial loss of information by the use of fractional factorial design. The P-B design is one of the classic examples for solving the above problem. In this, a quarter fractional design of five factors at two levels has (¼) x $2^5 = 8$ experiments, as in the present case. In the fractional factorial designs with four or more factors, the higher interaction terms can be ignored, so that only the main effects and two-factor interactions are evaluated.

The P-B design, often called the ruggedness test, has applications in validating methods that could be adopted as routine practice in laboratories. The factors with a high effect on the results are thus identified and can be subjected to a more rigorous investigation prior to validating the method. In collaborative trials, this design is particularly useful in giving prior warning to experiments that require careful control.

Since the experimental design is only a fraction of a complete factorial, each main effect is confounded with a large number of two-factor, three-factor and high-order interactions. However, since the selected design has n as an integral power of two, it is possible at least to identify the two-factor compounding.

If it is not required to study as many as (n-1) variables in n experiments, the unassigned variables are dummies, and no experimental condition is represented by + or – in the design, however the effects are calculated in the same way as those of the real variables. If no interaction and no experimental error occurs, the effect shown by the dummy variable would be zero. The non-zero results for the included dummy variables can be therefore used to estimate experimental variance. However, since it is impossible to determine whether the effect of a dummy variable is a measure of the uncertainty of the results, or whether it is actually due to one or more of the interactions with which it is confounded, the values of calculating significance in this way are questionable.

Since the design matrix is orthogonal, this estimate of error mean square of the linear effects agrees with the estimates produced with the least squares. In addition, the linear effects thus obtained are not confounded with one another and their estimates are unbiased. However, the P-B design can be extended to the analysis of non-linear multivariate regression equation with two significant two-factor interactions and more, as well.

In the present investigation, an attempt has been made at determining the relative importance of seven selected factors or variables (*viz*. dosages of china clay, talc, natural and precipitated calcium carbonates, binder, total solids of coating color and thickener) by the P-B design, in coating of base paper provided by various mills, and then at assessing the most pertinent variables (factors) influencing the properties of coated paper – namely: brightness, surface roughness, gloss, print gloss, printing picking velocity, print contact factor (m) and saturation ink density (D_∞). However, as the goal was to find out the relative effect of seven variables in terms of their preference for large-scale experiments, no attempt was made to develop linear multivariate regression equations with consequent evaluation of Student 't' and Fisher 'F' test. This procedure, not considering the development of multiple regression models, is in agreement with Broderic *et. al.*,[7] Cardwell[8] and Irvin *et. al.*[9]

Also, the above-mentioned seven process variables were relatively ranked, in accordance to their influence on different paper properties, to assess the most pertinent variable influencing the properties of coated paper. An indication on the relative importance of these factors would assist the design of effective laboratory-scale simulations of the commercial processes.

EXPERIMENTAL

A design matrix based on the Plackett-Burman design for eight experiments, involving seven variables, is given in Table 1. The assumption made was that, within the restricted range of each variable, the response is entirely linear. Table 2 shows a combination of process variables used for each coating experiment.

Thus, for coating experiment 1, the high levels of process variables A, B, C and E were used, as well as the low levels of variables D, F and G. The complete scheme is derived from this basic one-line design, which depends on the number of experiments. The subsequent lines are generated by the cyclic rotation of one space to the right (or left). The last line is always at low levels. When this is done, each variable will have an equal number of experiments – at high and low levels. Further examination shows that, when variable A is at its high level, B is high twice and low twice. Similarly, when A is at its low level, B is high twice and low twice. The same is true for the other variables. Thus, the net effect of changing the other variables cancels out, when A is calculated according to equation:

$$E_A = [\sum R, \text{Response at } (+)/(n/2)] - [\sum R, \text{Response at } (-)/(n/2)] \quad (4)$$

where E_A is the main effect of variable A and N is the number of results. The main effect of A is simply the difference between the mean values of the response at high and low levels, and the main effect of other variables can be calculated in the same way. Similarly, in the present case, the effect of changing factor, *e.g.* D, from low to high level (for N = 8) is given by (5):

$$D = \tfrac{1}{4}(R_2 + R_3 + R_4 + R_7) - \tfrac{1}{4}(R_1 + R_5 + R_6 + R_8)$$

Coating color preparation

Different coating pigments, collected from mills and suppliers, were evaluated for their particle size, particle size distribution and optical properties. The results are presented in Table 3.

Coating color formulations were prepared according to the experimental set-up, based on a P-B statistical design. Synthetically, the coating color preparation process occurs in the order given below. The parenthesis indicates the type of mixing in an either high- or low-speed mixer.

Pigments employed: (China clay, GCC, PCC/Talc) → distilled water (high speed mixing) → dispersant (high speed mixing) → defoamer (high speed mixing) → latex (low speed mixing) → thickener (low speed mixing) → NaOH (low speed mixing) → distilled water. The total solid contents of the coating slip were kept around 60%, and the pH of the coating slurry was maintained between 8 and 9.

Coating trials

A commercial base paper of 60 g/m^2, manufactured from the pulp of mixed hardwood and bamboo furnish, in a 80:20 ratio, was employed in all coating trials. All coating chemicals, *i.e.* pigment, latex, thickener, dispersant and deformer, were purchased from the coating plant itself. The viscosity of the coating color prepared was measured on a Brookfield viscometer. Water retention of the coating slip was measured using an AA-GWR gravimetric water retention meter (Finnish make model DT Paper Science, 1.5 bar, 120s, 5 μm filter).[10]

The coating trials were carried out on a laboratory bar coater (Zethner ZAA-2300) at a speed of 75 mm/s (4.5 m/min). The coated sheet was dried in a laboratory oven at a drying temperature of 117 °C, for 30 sec. The sheets were coated on one side with a coat weight of 20-21 g/m^2. The coated paper was then calendered in a laboratory calender using soft nip only. Each paper was calendered at a linear load of 100 kN/m, at a temperature of 60 °C, and a calendering speed kept at 20 m/min.

RESULTS AND DISCUSSION

Effect of process variables on the different characteristics of coated paper

Different sets of coating color based on the P-B design were applied to base sheets, then evaluated for different coated paper characteristics (Table 4). The main effects of the process variables on properties, such as viscosity, water retention, brightness, surface roughness, gloss, print gloss, picking velocity, print density parameter (m, D_∞) were evaluated and ranked accordingly (Table 5). The effect of the main influential variable, given as % of the mean, was also calculated (Table 6). The conclusion of the Plackett-Burman design results is discussed in the following paragraphs.

Optical characteristics

Brightness

The P-B design results reveal that, out of the pigments (*viz*. china clay, talc, natural and precipitated calcium carbonate), china clay induces negative effects, whereas other pigments have positive effects on coated paper brightness.

Table 1
Orthogonal design matrix of Plackett-Burman saturated design (N = 8; k = 7)

Exp. no.	Process variables						
	A	B	C	D	E	F	G
1	+	+	+	-	+	-	-
2	-	+	+	+	-	+	-
3	-	-	+	+	+	-	+
4	+	-	-	+	+	+	-
5	-	+	-	-	+	+	+
6	+	-	+	-	-	+	+
7	+	+	-	+	-	-	+
8	-	-	-	-	-	-	-

Table 2
Process variables used in experiments

S. no.	Process variable	Process variable conditions	
		Low level	High level
A	China clay (parts)	25	77
B	Talc (parts)	3	19
C	GCC (parts)	6	56
D	PCC (parts)	1	20
E	Binder (%)	10	18
F	Total solids (%)	50	65
G	Thickener (%)	0.2	1

Table 3
Coating pigments characteristics

Property	China clay	GCC	PCC	Talc
5%	<0.547µm	<0.605 µm	<1.55 µm	<3.185 µm
15%	<0.73 µm	<0.729 µm	<1.84 µm	<3.90 µm
35%	<1.19 µm	<0.896 µm	<2.26 µm	<4.81 µm
65%	<1.80 µm	<1.026 µm	<2.56 µm	<5.59 µm
Avg. particle size (µm)	4.18	1.32	3.26	7.09
Specific surface area (m^2/cm^3)	3.12	5.15	2.15	0.97
Brightness (%)	81.45	91.68	96.54	86.86
Whiteness (%)	71.25	86.50	93.21	76.48

Table 4
Coated paper characteristics obtained from experiments based on P-B design

Property	Brightness (%)	PPS roughness (µm)	Gloss (%)	Print gloss (%)	Picking velocity (m/s)	Density smoothness (m) (μ^{-1})	Print density (D_∞)	Viscosity (mPas)	Water retention (g/m^2)
Exp. 1	80.0	1.77	41.4	73.5	225	0.34	2.42	50	2.24
Exp. 2	82.6	1.64	40.6	78.0	90	0.31	2.41	117	1.58
Exp. 3	82.4	1.92	38.2	75.8	240	0.30	2.43	57.4	1.48
Exp. 4	76.3	1.30	55.5	88.3	180	0.33	2.30	138	1.30
Exp. 5	77.5	1.41	40.0	84.8	276	0.30	2.38	450	1.38
Exp. 6	80.3	1.23	50.5	81.0	120	0.31	2.54	322	1.05
Exp. 7	78.4	1.07	44.0	78.7	93	0.24	2.91	402	0.95
Exp. 8	79.7	1.27	62.0	82.2	67	0.18	3.42	456	2.03
Mean	79.65	1.45	46.5	80.28	161.3	0.288	2.60	249.05	1.50

Table 5
Main effects of process variables on both properties and relative ranking (based on P-B design)

Variables	Brightness (%)	PPS roughness (µm)	Gloss (%)	Print gloss (%)	Picking velocity (m/s)	Density smoothness (m) (μ^{-1})	Print density (D_∞)	Viscosity (mPas)	Water retention (g/m^2)
China clay	-1.80 (2)	-0.22 (3)	7.15 (3)	0.27 (6)	40.25 (3)	0.03 (2)	-0.11 (5)	-42.1 (5)	-0.23 (4)
Talc	0.5 (6)	0.04 (7)	-10.05 (1)	-3.07 (3)	12.75 (6)	0.01 (3)	-0.12 (4)	11.4 (7)	-0.07 (7)
GCC	3.3 (1)	0.37 (1)	-7.7 (2)	-6.4 (1)	14.75 (5)	0.05 (1)	0.30 (3)	-344.9 (1)	0.17 (6)
PCC	0.55 (5)	0.06 (6)	-3.9 (7)	-0.2 (7)	-21.2 (4)	0.01 (3)	-0.18 (6)	-140.9 (4)	0.035 (3)
Binder	-1.2 (3)	0.300 (2)	5.5 (5)	0.63 (4)	137.8 (1)	0.05 (1)	-0.43 (1)	-150.4 (3)	0.20 (5)
Total solids	-0.95 (4)	-0.112 (4)	4.125 (6)	5.4 (2)	10.3 (7)	0.05 (1)	-0.38 (2)	15.4 (6)	-0.41 (2)
Thickener	0.1 (6)	-0.11 (5)	-6.7 (4)	-0.4 (5)	41.7 (2)	0.01 (3)	-0.07 (6)	182.4 (2)	-0.63 (1)

Table 6
Main effect as percentage of mean values (based on P-B design)

Variables	Brightness (%)	PPS roughness (µm)	Gloss (%)	Print gloss (%)	Picking velocity (m/s)	Density smoothness (m) (μ^{-1})	Print density (D_∞)	Viscosity (mPas)	Water retention (g/m^2)
China clay	-2.25	-15.17	15.3	0.34	-24.9	10.3	-4.2	-17.0	-15.4
Talc	0.62	2.75	-21.6	-3.82	7.9	3.4	-4.6	4.5	-4.72
GCC	4.14	25.5	-16.5	-7.9	9.14	17.2	11.4	-138.5	11.0
PCC	0.70	4.22	-8.4	-0.25	-13.2	3.45	-6.9	-56.58	12.2
Binder	-1.5	20.7	11.8	0.78	85.4	17.2	-16.5	-60.4	13.0
Total solids	-1.2	-7.72	8.8	6.7	6.4	17.3	-14.6	6.18	-24.4
Thickener	0.12	-6.9	-14.4	-0.5	25.88	3.45	-2.72	73.2	-38.5

In the experiments performed, the binder and total solids of the coating color showed negative effects, whereas the thickener used to improve water retention had a positive effect.

The main effect, expressed as percent of the mean value, reveals the highest effect of natural calcium carbonate (4%), followed by china clay (2.25%) and total solids (1.2%).

The analysis of the different optical properties of the coating pigments also indicates that the precipitated calcium carbonate used in the above experiments show the highest brightness and whiteness, followed by ground calcium carbonate, talc and china clay – which explains the brightness value of coated paper.

Gloss

China clay had a positive effect on gloss, whereas a negative effect was observed in the case of talc, ground and precipitated calcium carbonates. Talc gave the highest negative effect (21.6% of the mean effect), followed by ground calcium carbonate (16.5%) and precipitated calcium carbonate (8.4%), indicating that the introduction of these pigments with china clay will give coated paper with a somewhat matt finish. The higher surface gloss in case of china clay (15.3%) is probably due to the relatively lower void volume formation in the coating layer.[8,9] The binder level (11.8%) and the total solid of coating color gave a positive effect on gloss. The results obtained agree with the findings of Gron[11] and Hagymassy.[12]

Surface characteristics

Surface roughness

China clay had a negative effect on surface roughness, *i.e.* a smoother surface is obtained when china clay is used as a pigment in coating (15.2% of the mean effect). The positive effects of talc and precipitated calcium carbonate were comparable (3.0 and 4.2%), being higher for ground calcium carbonate (25.5% of the mean), indicating that the ground calcium carbonate containing coated paper will be comparatively less smooth. Talc was able to enhance the evenness of the paper surface, probably due to the fact that talc particles are flat in shape and possess a high aspect ratio. The binder level had a positive effect, of the 20.7% order, whereas the total solids and the thickener gave negative effects (7.7 and 6.9%, respectively).

Printing characteristics

Print gloss

Print gloss was improved by the presence of china clay and by its positive effect. The highest negative effect was induced by ground calcium carbonate (7.9%), followed by talc (3.8%) and precipitated calcium carbonate (0.25%). The binder level and the total solids showed positive effects (0.8% and 6.7%), whereas the thickener (0.5%) had a negative effect.

Picking velocity

The presence of talc and ground calcium carbonate improved picking velocity, while china clay and precipitated calcium carbonate caused a negative effect. The binder level, total solids and thickener showed positive effects on picking velocity. Out of all variables, the binder level showed its maximum effect as a % of the mean (84.5%), followed by thickener (25%) and PCC (13.8%).

Print density

The print density curve, *i.e.* the optical density of print plotted against the ink film thickness on the printing form gives indication about the ink requirement of paper, for attaining a particular print density. In the print density curve, density D (*i.e.* contrast with respect to unprinted paper) is plotted against ink film thickness. Such a density curve generally follows the expression:[13]

$$D = D_\infty (1 - e^{-mx})$$

where D_∞ is saturation density, *i.e.* the density value approached by the theoretical curves if ink film thickness is increased indefinitely, m is the contact factor or density smoothness, showing how quickly saturation density is obtained, and x is ink layer thickness.

The ground calcium carbonate pigment shows its highest effect on the contact factor, m (*i.e.* 11.4%), followed by precipitated calcium carbonate (6.9%), talc (4.6%) and china clay (4.2%). This indicates that ground calcium carbonate helps improve print density more than the other studied pigments do, having also a positive effect on D_∞. The other pigments, like china clay, talc and precipitated calcium carbonate, had negative effects on D_∞. The binder level, total solids and thickener had a positive effect on m, and a negative one on D_∞.

Use of talc in coating formulation

The introduction of talc should be encouraged in matt finish or semi-matt finish coated papers subsequently offset-printed, as the following features of the offset process are especially demanding for talcs:

- Tacky ink emulsified by moisturizing solution

- Hydrophilic moisturizing solution
- Offset blanket materials.

Talc helps reduce the abrasivity of matt paper surface and the tendency of ink to scuff or rub off. Ink scuff occurs when the ink paper surface degrades the ink layer. With less abrasive surfaces, the ink layer resists better.

The coarse ground calcium carbonate generally used for matt papers creates an abrasive surface against ink that can be controlled by using talc in the recipe. Compared to a 100% carbonate recipe, talc improves the ink holdout. Viscosity is positively affected by total solids and thickener. Ink setting/ink holdout, especially in offset printing, is a function of ink penetration and/or ink absorption into the coating layer. The solvent of the printing ink should obviously escape from the ink after printing. The more dried ink is retained on the outer surface of the paper, the higher will be ink density or, accordingly, the lower will be the ink demand.[14] If the pores of the coating layer are too large, obviously the printing ink will penetrate easily the paper and the print gloss will be low, which is the case observed for ground calcium carbonate. To avoid this, ground and precipitated calcium carbonate should be preferably used, along with platy pigments – like clay or talc.

Effect on rheological properties of coating color

The effects that coating color, made from different combinations of pigments and other additives, may have on rheological properties were studied by measuring the viscosity and water retention value of different formulations (Table 4). The results obtained show that, out of the different pigments used, talc had a positive effect on coating color viscosity, whereas all other pigments had negative effects. The highest negative effect was shown by precipitated calcium carbonate, indicating that the introduction of excess precipitated calcium carbonate will change the rheological characteristics of the coating color. This is probably due to the high aspect ratio and narrow particle size distribution of precipitated calcium carbonate, which may affect its ability to be pumped, and also to the flow behavior in coating equipments.

The water retention value is negatively affected by china clay and talc, indicating that these pigments help in improving the water retained by the coating layer, whereas the water retention value (WRV) is positively affected by the presence of ground and precipitated calcium carbonates. The positive water retention value means that less water is retained by coating color on the paper surface.

To get better coating, the pigment should assure good water retention of the coating color. If water is not retained to some extent, it will be instantly drawn into the paper during application, and a hard cake will be built up on the metering device. The water retained is worst with ground and precipitated calcium carbonates, comparatively with china clay, probably due to the fact that, in the case of china clay and talc, water simply has to go a longer way through a tortuous platy packing while, in the case of ground and precipitated calcium carbonates, particle size distribution is narrower, so that water has a shorter penetration path and a lower tortuousity factor.

CONCLUSIONS

The Plackett-Burman design serves to ranking a large number of process variables according to their relative importance. The relative effect of the different process variables studied by the P-B model indicates that:

- China clay has a negative effect on the brightness of coated papers, compared to other pigments – such as ground and precipitated calcium carbonate and talc.
- China clay has a positive effect on gloss, while talc, ground and precipitated calcium carbonate give negative effects. Ground calcium carbonate shows the highest negative effect on gloss. The binder level and the total solids of coating color had positive effects on gloss.
- A smoother surface is produced when China clay is used as a pigment, followed by talc and precipitated calcium carbonate.
- China clay has a more positive effect on print gloss, while ground calcium carbonate has a negative effect. Binder level and total solids show positive effects, while the thickener had a negative effect on print gloss.
- China clay and precipitated calcium carbonate had negative effects on

picking velocity, while talc and ground calcium carbonate improve it.

- Ground calcium carbonate helps improve print density more than other pigments do. It also has a positive effect on D_∞ (saturation density). The binder level, total solids and thickener have a positive effect on m (contact factor or density smoothness), and a negative one on D_∞ (saturation density).
- The introduction of talc should be encouraged in matt finish or semi-matt finish coated paper for offset printing.
- The water retention value (WRV) is negatively affected by china clay and talc, indicating that these pigments improve the water retained by the coating layer, being positively affected by ground and precipitated calcium carbonate.

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