JPMTR 1411 Professional communication DOI 10.14622/JPMTR-1411 Received: 2014-07-04 UDC 655.1 : 762-035.67 | 66-97 Accepted: 2014-12-06

Analysis of the thermal behavior of gravure inks: comparing experimental results and numerical methods

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Abstract

Gravure inks are frequently applied in multiple layers as prints on packaging material. Besides giving some information about the product, the printed ink film can be used also for protection. The compound printed layer of different colored gravure inks (of equal thickness) plays an important role in protecting the product contained within the package from external atmospheric conditions. For example, heat transfer either from the outside or the inside of the package can be controlled by a properly designed ink film combination on the package. The color inks generally used are specialty white, standard yellow, magenta and cyan, together with black, applied consecutively. If these ink coatings are not long-lasting, or the system is not properly designed, this may lead to failure to protect the packaged product due to unwanted transfer. In the present investigation, the thermal properties of free films of different colored inks were measured individually and also in combination to assist in the design of appropriate ink layers on packaging leading to extended shelf-life for the product. The measured thermal properties are thermal conductivity, specific heat and the coefficient of linear expansion. Further, the thermal contact resistances of different color combinations, contact surface temperatures and thermal stress developed in the coating were determined using an algebraic method. A finite element method was also adopted for determining the contact surface temperature and thermal stresses developed. These numerical results were compared with the experimental findings. It was found that the heat transfer rate either from the package or from the outside will be much lower if the thicknesses of all the ink layers are equal. Also, it is proposed that if the coatings of inks are printed in the order of WKCMY, the protection of the product will be optimal.

Keywords: coating, coefficient of linear expansion, thermal conductivity, specific heat, contact thermal resistance

1. Introduction

Packaging is the most widely applied technique for enclosing or protecting products for distribution, storage, communication, sale and use. The materials used for packaging purposes are either metallic or non-metallic or a combination of both. Besides the structural packaging material itself, the use of gravure ink as a functional coating in the form of different colors of equal thickness applied onto packaging has an important role in protecting the product contained from the external atmosphere, with particular emphasis on preventing heat transfer either from the outside or from the inside of the package. Figure 1 shows the geometry of a package structure consisting of five different colored ink layers applied with equal thickness on the packaging base. The thicknesses of the different colored ink layers were made equal since these are printed using the CMYK model along with a white background using a gravure printing process.

To serve the purpose of protection, the package as well as the films of ink must be dimensionally stable under different ambient conditions during the life cycle of the product. So, the thermal and mechanical properties of the films, made of different colored inks (individual and combined ink films), will play a vital role in protective coatings (Paul, Naskar and Pal, 2011).

Figure 1: Geometry of a package structure printed with the CMYK model along with a white background (WCMYK)

2. Experimental procedures

2.1 Ink films and sample preparation

For the measurement of various thermal properties of gravure inks, free films of five different colored inks were made using a special technique developed earlier (Paul et al., 2002). The free films consist of individual as well as of combinations of different colored inks. Measurements of the thermal properties of the different colored inks are limited to thermal conductivity, specific heat and the coefficient of linear expansion. The thermal conductivity of different combined ink films was also measured to find out the contact thermal resistance of the ink combinations.

The sample preparation technique developed earlier (Paul et al., 2002) consists of two steps: (a) formation of uniform sample layers of the coatings, and (b) separation of the adherent films. A metal plate, of dimensions equal to or greater than that of the sample, was taken at first. A uniform layer of bee wax was coated onto the plate. A gravure ink layer was applied to the wax surface by roller. The ink layer was allowed to dry sufficiently before giving a second layer onto it. This layering process was continued only after the previous layer was fully dried and this process was continued until the required thickness was obtained. The ink film was carefully separated from the waxed metal plate by a sharp knife. This procedure leaves the film alone. Several samples of different thickness, each made up of cyan, magenta, yellow, black and white colored gravure inks, were used for the present studies. Combined ink films were made by applying ink layers on already made free films of ink.

2.2 Thermal conductivity

The thermal conductivity of the sample was measured as per ASTM C-177-45 (ASTM International, 1945). The sample was placed inside a guard plate heat conductivity unit (Model DTI-12, S. C. Dey & Co. Ltd., India) as shown in Figure 2. The sample was then heated over a certain temperature range at different ambient conditions and the temperatures on both sides of the sample were measured using thermocouples. The details of the measurement procedure are given by Paul et al. (2004a). For the measurement of the thermal conductivity of ink, about 150 data points, each of five different colors of different thickness ink films, were considered.

2.3 Specific heat

The specific heat was also measured using the guard plate heat conductivity unit as shown in Figure 2. When one side of a sample is heated, the layer adjacent to the heater absorbs part of heat, leading to an increase in its own temperature. A part of the heat applied is lost by way of radiation from the surface and convection through the air. The remainder of the heat available passes into the adjacent layer, increasing the temperature of that layer. The instrument has been fabricated in such a way as to minimize the losses due to radiation and convection. Details of the measurement procedure are given in Paul et al. (2004a). In order to measure the specific heat of the samples, about 150 data points were taken into consideration for each of five different colored samples. The mean values of the specific heat of the samples are shown in Table 1.

2.4 Coefficient of linear expansion

The coefficients of linear expansion of free films of inks of different colors were measured using a resistance type strain gauge. Strain gauges having a gauge factor of 2.0 and an electrical resistance of 350 Ω were used. In order to measure the coefficient of linear expansion of the sample, the strain gauge was fixed on the surface of the sample. The strain gauge was fixed to the center of the sample using a thin layer of standard adhesive. The coefficient of linear expansion of the sample may be affected if the thickness of the adhesive is large. To compensate for this effect, a thick sample was taken for measurement and the experiment was repeated several times. The sample and

strain gauge combination were placed in a closed chamber, the temperature of which was varied over a wide range from sub zero Celsius to room temperature by applying liquid nitrogen. The surface temperature of the sample was measured using a Hewlett Packard Data Acquisition System (Model 34970A) through a 'K type' thermocouple. The experimental arrangement for such measurement has been published by Paul, Naskar and Pal (2011). Details of the measurement procedure are given by Paul et al. (2004a). For the measurement of the coefficient of linear expansion of free films of five different colored ink samples, nearly 250 data points for each were used. The mean values thus obtained are shown in Table 1.

2.5 Contact thermal resistance

Thermal contact conductance is the measure of heat conduction between different solid bodies in thermal contact. The thermal contact conductance coefficient, h_c , is a property indicating the thermal conductivity, or ability to conduct heat, between two bodies in contact. The inverse of this property is called contact thermal resistance.

The contact thermal resistance R_c has been calculated using Equation 1 (Chapman, 1984; Holman, 1997) assuming that heat is flowing uniformly from layer 1 to layer 2 and that all the ink layers are isotropic in nature.

$$
R_c = \frac{1}{b_c} = \left(\frac{(\theta_1 - \theta_2)A}{Q}\right) - \left(\frac{l_1}{K_1} + \frac{l_2}{K_2}\right)
$$
 [1]

where h_c is the thermal conductance, l_1 the thickness of the hotter body in m, l_2 the thickness of the colder body in m, K_1 and K_2 the conductivities of layers 1 and 2, respectively, in Wm⁻¹K⁻¹, ($\theta_1 - \theta_2$) the temperature difference between the upper and lower surfaces of the two layers in K, A the area of the layers in m², and Q the heat rate (power) in W.

The temperatures θ_1 and θ_2 were measured experimentally using the guard plate heat conductivity unit. In order to measure θ_1 and θ_2 , a block of two different colored gravure ink layers was made by applying the second color on the top of first color. The color combination was selected according to Figure 1.

2.6 Contact surface temperature

2.6.1 Algebraic method for calculation of contact surface temperature

When two bodies are in contact, the temperature at the contacting surface is called contact surface temperature.

For a two layer system, assuming unit layer area, when the upper and lower surface temperatures are T_1 and T_3 , and T_2 is the contact surface temperature, T_2 and T_3 can be calculated using Equations 2 and 3

$$
T_2 = \frac{\left[T_1 + \left\{ (l_1 / K_1 + R_c) / (l_2 / K_2) \right\} T_3 \right]}{\left[\left\{ (l_1 / K_1 + R_c) / (l_2 / K_2) \right\} + 1 \right]}
$$
\n^[2]

$$
T_3 = T_1 - \{ (Q/A) (R_c + l_1/K_1 + l_2/K_2) \}
$$
\n⁽³⁾

where R_c is the contact thermal resistance, and l_1 and l_2 the thicknesses of the two consecutive layers in m.

The assumption underlying this calculation is that there is no energy loss in the system. The contact thermal resistance of the system of coating layers WCMYK is calculated progressively using a successive two layer system. The values thus obtained are utilized to find the thermal stresses developed using an algebraic method.

2.6.2Determination of contact surface temperature by finite element method

In order to calculate the contact surface temperature of the printed package with five layers of different inks, the differential Equation 4 (Akin, 1986) has been adopted for each individual layer and applied in the finite element method (FEM).

$$
K\frac{\mathrm{d}^2 T}{\mathrm{d}x^2} + Q = 0\tag{4}
$$

The finite element solution for the above equation is used as in Equation 5

$$
K^{\rm e}T^{\rm e} = \mathcal{Q}^{\rm e} \tag{5}
$$

where $K^e = \frac{AK}{l}$ 1 1 1 -1 1 − is the element stiffness matrix,

 T^e the nodal temperatures in K, and Q^e the nodal heat flux in W per unit time.

Assembling layerwise homogeneous elements that have different conductivities, a non-homogeneous, one dimensional model has been considered. Steady state heat conduction through unit area is assumed for all elements. The system considered consists of six nodes and five elements, since the coating layers here are five, namely white, cyan, yellow, magenta, black ink layers, and the ink layers are isometric in nature. The values thus obtained were used to find out the thermal stresses developed.

2.7 Thermal stress

2.7.1 Definition

Thermal expansion or contraction cannot occur freely in all directions of any solid body because of geometry, external constraints or the existence of temperature gradients. So, stresses are produced. Such stresses caused by a temperature change are known as thermal stress. Thermal stress occurs as a result of a non-uniform distribution of temperature in different parts of the body.

2.7.2 Algebraic method for calculating thermal stress

In order to calculate thermal stress, Equation 6 was used

$$
\sigma = E\alpha \Delta T \tag{6}
$$

where *σ* is the stress in Pa, *E* the Young's modulus in Pa, α the coefficient of linear expansion, and ΔT the temperature difference between the two planar surfaces in K.

The modulus of elasticity (*E*) of free films of different colored inks was measured using a special technique developed earlier (Paul et al., 2004b). To validate the values obtained by the algebraic method, the finite element method was used.

2.7.3 Determination of thermal stress by finite element method

The finite element temperature distribution is expressed according to Equation 7 (Buchanan and Rudramoorthy, 2006) – see Figure 3,

$$
T = [N] \{ u(t) \}
$$
\n
$$
(7)
$$

where [*N*] is the element stiffness matrix and $\{u(t)\}\$ the nodal temperatures at time *t*.

The thermal load due to temperature difference can be expressed as in Equation 8

$$
Q^{\rm e} = \frac{EA/\alpha \Delta T}{(x_2 - x_1)} \begin{cases} -1 \\ +1 \end{cases}
$$
 [8]

where E is the effective Young's modulus of the coating in Pa, A the cross sectional area of the layer in m², *l* the layer thickness in m, and (x_2-x_1) the nodal distance in m.

The thickness *l* and nodal distance (x_2-x_1) are both on the same scale and in our case equal to 0.03 mm. For steady state and one dimensional heat conduction, the stress developed on a unit area of an individual layer can be expressed as in Equation 9.

$$
\sigma = \frac{E}{(x_2 - x_1)}[-1 \ 1]u - E\alpha \Delta T \tag{9}
$$

where σ is the stress on individual elements.

The corresponding nodal representation of the FEM is shown in Figure 3.

3. Results

Our measurements were conducted on free films of gravure inks of individual cyan, magenta, yellow, black and white, and different color ink layers in combinations of two colors, namely W-C, C-M, M-Y and Y-K. Table 1 shows the results (mean values) of the measurements of thermal properties of the samples as described in 2.2 , 2.3 and 2.4.

Gravure ink film	Thermal conductivity $(W m^{-1} K^{-1})$	Specific heat $(kJ kg^{-1} K^{-1})$	Coefficient of linear expansion, α (K ⁻¹) · 10 ⁻⁶
Cyan ink	0.1307 ± 0.04	0.1028 ± 0.17	1.3217 ± 0.18
Magenta ink	0.1399 ± 0.01	0.1102 ± 0.18	1.4984 ± 0.11
Yellow ink	0.1391 ± 0.03	0.0885 ± 0.18	1.5631 ± 0.79
Black ink	0.2488 ± 0.05	0.1331 ± 0.27	1.0695 ± 0.17
White ink	0.1537 ± 0.02	0.1203 ± 0.25	0.3628 ± 0.25

Table 1: Thermal properties of different gravure inks

Figures 4–8 show the thermal conductivity as a function of temperature for the different samples under different atmospheric conditions, and Figures 9–13 show the variation of the coefficient of linear expansion with temperature under different atmospheric conditions.

Heat flow is directly related to the thermal conductivities of the bodies in contact. The conductivity of ink films in combination was measured in order to determine the contact (skin) thermal resistance (average calculated values are shown in Table 2).

The measured values of *E* of different colored gravure inks are given in Appendix A. Table 3 shows the calculated values of thermal stress for the model (Figure 3) when heat is transferring from outside to inside of the package for a temperature difference of 40 °C.

	Thermal stress (MPa)	
Individual color gravure ink	Algebraic	FEM
Layer $5(K)$	0.126	0.139
Layer $4(Y)$	0.704	0.716
Layer $3(M)$	1.623	1.629
Layer $2(C)$	0.870	0.872
Laver 1 (W)	0.157	0.157

Table 3: Thermal stress for our model when heat is transferring from outside to inside of the package

Table 4 shows the calculated values of thermal stress for the model (Figure 3) when heat is transferring from inside to outside of the package for the same temperature difference as previously.

Individual color	Thermal stress (MPa)	
gravure ink	Algebraic	FEM
Layer 1 (W)	0.157	1.233
Layer $2(C)$	0.870	1.949
Layer $3 \, (M)$	1.623	2.706
Layer $4(Y)$	0.704	0.829
Layer $5(K)$	0.126	0.252

Table 4: Thermal stress for the model when heat is transferring from inside to outside of the package

4. Analysis

Figures 4 and 5 show that the thermal conductivity of the white and cyan color ink layers decreases with temperature. The relative humidity of the atmosphere as well as the ambient temperature influence the thermal conductivity of both inks. For the white ink layer, the higher the ambient temperature and relative humidity, the greater the thermal conductivity. This implies that the heat conduction rate decreases with an increase in temperature.

 0.2

Figure 4: Variation of thermal conductivity of free film of white colored gravure ink with temperature at different atmospheric conditions

Figure 5: Variation of thermal conductivity of free film of cyan colored gravure ink with temperature at different atmospheric conditions

Figure 6: Variation of thermal conductivity of free film of magenta colored gravure ink with temperature at different atmospheric conditions

Figure 8: Variation of thermal conductivity of free film of black colored gravure ink with temperature at different atmospheric conditions

Figure 9: Variation of coefficient of linear expansion of free film of white colored gravure ink with temperature at different atmospheric conditions

Figure 10: Variation of coefficient of linear expansion of free film of cyan colored gravure ink with temperature at different atmospheric conditions

From the thermal conductivity curve of the magenta ink film (Figure 6), it can be observed that the thermal conductivity remains more or less constant up to the temperature of 52 °C and decreases thereafter. The relative humidity also influences the thermal conductivity of the same. In this case, the effect of relative humidity is the reverse. The thermal conductivity decreases with an increase in relative humidity at least up to 52 °C as shown. This may be due to the type of pigment used in magenta ink. The pigment used in the magenta ink used is Barium 2B red toner (see Appendix B). This pigment has a tendency to release water soluble azo coupling groups as well as to absorb water. The strange behavior of the magenta ink film at different relative humidities and at lower temperature is due to the absorbed moisture by the pigment, which later evaporates.

For the yellow color ink film, the thermal conductivity decreases exponentially with an increase in temperature (Figure 7). For black ink film, the thermal conductivity decreases with the increase in temperature (Figure 8). The relative humidity and atmospheric temperature greatly influence the thermal conductivity of both inks. For black ink, it is obvious that the higher the temperature and relative humidity, the greater the thermal conductivity.

For the white ink film, the coefficient of linear expansion is at its maximum in the subzero range (i.e., −28 °C to −20 °C) and thereafter it decreases sharply with an increase in temperature (Figure 9). This may be due to the higher quantity of metallic oxide present in the white ink (see Appendix B) compared to the non-metallic organic part. As a result, the exchange of heat between the pigment and other material starts much earlier than when at a normal ambient temperature. For the cyan ink film, it can be observed that the coefficient of linear expansion is at its maximum at about −10 °C and decreases thereafter (Figure 10). The higher the ambient temperature and relative humidity, the lower the expansion coefficient of the cyan ink film. For magenta ink film, the expansion has a maximum at close to $+7$ °C for both ambient conditions (Figure 11). For both cyan and magenta ink films, it can be observed that the relative humidity and the temperature influence the coefficient of linear expansion. The higher the relative humidity and the ambient temperature, the lower the coefficient of expansion of magenta ink film within a normally applicable temperature range. For yellow ink film, it is seen that the coefficient of expansion decreases in the subzero (Celsius) range (i.e., −17 °C to −1 °C) and after that it remains constant in all ambient conditions (Figure 12). This may be due to the lower quantity of pigment present in comparison to binder (see Appendix B) in the yellow ink. It can also be observed that the ambient temperature influences the coefficient of linear expansion: the higher the ambient temperature, the higher the coefficient of linear expansion. For black ink film, it can be observed that coefficient of linear expansion increases with increasing temperature and has a maximum at a temperature range of −27 °C to −10 °C while remaining constant at higher temperatures. This may be due to the higher quantity of pigment present in the dried ink film in comparison to other materials such as binder, additives, etc. (see Appendix B). As a result, heat exchange between the pigment and the other materials starts much earlier than at 0 °C.

Tables 3 and 4 show the changes in numerical stress within the layer as heat is transferring from one layer to another. It can be seen that when heat is transferring from outside to inside as well as from inside to outside the maximum stress generation takes place in the magenta layer and the minimum in the black layer.

5. Conclusions

It has been found that the thermal conductivity of the black ink film is greater than the thermal conductivities of the free films of yellow, magenta and cyan color inks, which are more or less on the same level. The thermal conductivity of the white background layer is slightly higher than that of the free films of yellow, magenta and cyan colored inks. The coefficient of linear expansion of the free film of white ink is much lower. It has also been observed that the skin contact thermal resistance of a white-cyan combination and a cyan-magenta combination are more or less the same whereas the skin contact thermal resistance of a magenta-yellow ink combination and a yellow-black ink combination are lower than that of the previous two layers. This implies that the heat transfer either from the package or from the outside will be much lower if the thicknesses of the ink layers are equal. Again, the thermal stresses will be high at the contact point of a white and cyan layer due to differential thermal expansion of the two different layers. This may lead to cracking in the white layer due to its modulus of elasticity being smaller than that of cyan.

The thermal stress developed in a cyan layer is higher than that of a white layer. This is in accordance with the experimental findings.

With respect to the modulus of elasticity, ink films may be represented by M>C>Y>W>K. As per thermal stress analysis is concerned, when the package is subjected to external heating due to changes in ambient conditions, the thermal stress developed on the different ink films may be represented as M>C>Y>W>K.

Based on the measurements of thermal and elastic properties and on the numerical analysis, it is proposed that the coating of inks should be printed in the order of WKCMY to give better protection of the product by preventing heat transfer as well as failures of the coating layers.

White is used as a background layer to get the effect of the substrate as well as the effect of multicolor printing. Moreover, white ink uses titanium dioxide (anatase form) which has the property of preventing the passage of UV light. Regarding thermal conductivity, the ink films can be represented by K>W>M>Y>C and when considering the coefficient of linear expansion, the ink films may be represented by Y>M>C>K>W. This implies that expansion is the smallest on the white layer and black exhibits the least stress generation as per stress values. For these above reasons, the longevity of the packaging as well as the heat transfer rate from the outside will be optimal if the layers are printed in the order of WKCMY.

Finally, standard inks are considered as insulating material for cost reasons as well as a way to get proper superimposed color printing effect. Moreover, the white ink layer has the property of UV protection of the product since it is printed in solid form as a background layer on the package.

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Appendix A: Measuring the modulus of elasticity of free films of different colored gravure inks

Firstly, free films of different colored gravure inks have been prepared. Then a resistance type strain gauge having gauge factor of 2 is attached onto the surface of the ink film using standard adhesive. Thereafter the sample is hung on a horizontally fixed frame (Paul et al., 2004a) and it is subjected to different loads with the strain gauges connected to the data acquisition system to collect strain data at different loading conditions. The modulus of elasticity is then calculated.

Appendix B: Compositions of the different colored gravure inks

Liquid gravure ink contains three major ingredients together with smaller quantities of additives. The major ingredients are pigments (including dyestuff), binder, and solvent. The additives used consist of materials such as driers, plasticizer, wax, etc.

The composition of the gravure inks used in this work is as follows:

Black colored ink:

The pigment present in the black colored gravure ink used is carbon black (CI Pigment Black 7 no. 77266) and the vehicles used in the ink are ethanol and ethyl acetate (Leach et al., 1988). The pigment/binder ratio in the gravure ink used is 2.308.

Magenta colored ink:

The pigment present in the magenta colored gravure ink used is barium 2B red toner (CI Pigment Red 48.1) and the vehicles used in the ink are ethyl acetate and n-propyl acetate (Leach et al., 1988). The pigment/binder ratio in the gravure ink used is 0.9.

Yellow colored ink:

The pigment present in the yellow colored gravure ink used is diarylide yellow (CI Pigment Yellow 12) and the vehicles used in the ink are ethanol and ethyl acetate (Leach et al., 1988). The pigment/binder ratio in the gravure ink used is 0.5.

Ingredients by weight Phthalocyanine blue 12.0 Alcohol soluble polyamide resin 13.0 Nitrocellulose 18.0 Dioctylphthalate 2.0 Antioxidant 0.5 Ethanol 40.0 Ethyl acetate 7.5 n-propyl acetate 5.0 Amide wax. 1.0 Polyethylene wax 1.0

The pigment present in the cyan colored gravure ink used is phthalocyanine blue (PBI 15.3) and the vehicles used in the ink are ethanol, ethyl acetate and n-propyl acetate (Leach et al., 1988). The pigment/binder ratio in the gravure ink used is 0.387.

White colored ink:

The pigment present in the white colored gravure ink used is titanium dioxide (CI Pigment White 6) and the vehicles used in the ink are ethanol and ethyl acetate (Leach et al., 1988). The pigment/binder ratio in the gravure ink used is 2.727.

Cyan colored ink: