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# The effect Ecofriendly Super hydrophobic Nano composites Coating and Penetration of Printing Inks on Paperboard Packaging Materials

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**Abstract:**
The packaging industry is one of the pioneering industries facing many challenges during the production cycle. Migration of printing ink in packing items is the harmful problem for customer, Therefore, this study aims to reduce the penetration of ink into the product, one of the unavailable technologies is coating printing substrates with a polymer layer. Ecofriendly Super hydrophobic coating was prepared in aqueous medium based on silicon dioxide and characterized with particle size analyzer Dynamic laser scattering (DLS) and zeta potential. Paper duplex 180 g/m$^2$ were coated with prepared superhydrophobic Nano coating through spray coating with emulsion solution. The coated paper sheets were printed by lithooffist and testes by X-rite instrument. This polymer can seal the pores of the printing materials. It is also a barrier to prevent the leakage and deportation of ink particles into the packaging that can occur in various raw materials, especially untreated materials as well as. One of the advantages of this polymer is to reduce the penetration of ink consumed for printing, that is, it is of double benefit, which is protection and reduce ink penetration, and the research results show the effective use of this polymer layer, whether in roughness, water absorption, Ink penetration and printing properties

**Keywords:**
Superhydrophobic (SH),
Coating,
Paperboard,
Penetration,
Packaging Materials

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## 1. Introduction

Packaging materials is one of a superior feeding industry for multidiscipline applications in various food and non-food industries (Ramin Jouneghani, 2020). Packaging materials vary from paper, Kraft bags, aluminum paper, cellophane, carton, plastics of all kinds (Jinwu Wang, 2018). Paperboard is the most applied packaging materials in first and/or second packaging. Naturally paper and paperboard packaging materials have multi size pores with wide dimeters which consider as one of disadvantage especially in printing ink consuming (ana Balea, 2019). Hence the idea of research came to increase the value of the package by coating with an ecofriendly nanocomposite layer as superhydrophobic coating before printing the package. The most important role of coating is that it is a pore filler to enhance the topographic character of the coated surface (V. Jothi, 2020). Superhydrophobic nanocoating was selected to applied over paperboard surface to improve the surface energy and morphological structure after coating (S. Ibrahim, 2017). Printing of paperboard can be applied with different technologies like, flexographic, gravure and lithooffset. Coated and uncoated paperboard (duplex 180 g/m$^2$) were printed with Lithoffset. In this research, duplex paper has been used in Practical tests as it is one of the most popular packaging materials, especially in bakery packaging.

### Originality of the work:

This research work is related to one of high value industrial, packaging materials, as feeding industry for food and non-food packing products. Modification of packaging materials surface with novel super hydrophobic coating has a great impact to enhance the printability and visual behavior of printing packaging materials.

### Research Problems:

1- Penetration of ink components into the package.
2- Most usable coating are solvent base polymer coating

### Research objectives:

1. Reduced the migration of some ink components to the packing product.
2. Study the printing properties of packaging products to apply a superhydrophobic nano coating layer before printing
3. Reduce the risk of print inks penetration of printing inks into the packing product.

### Research Importance

The packaging industry has an important vital role as feeding industry for many industries. The packaged products must be prevented from interacting with printed packaging materials because one of the basically of packaging to save items without hazard material transferring. Therefore, the idea of the research based on finding nanotechnology solutions to solve the migration problem of some ink components into the packing product.

## Literature Review:

**Smart self-cleaning superhydrophobic nanocoating**

Most of available marketing coatings based on non-aqueous solvents which restricted its applications without environmental hazards are available dioxide nanoparticles were encapsulated in polystyrene through miniemulsion polymerization techniques as a cheap and easy applicable coating (S. Ibrahim, 2009).

Superhydrophobic Nano coating is special coatings with desirable properties like super hydrophobicity, play an essential role in development of coated substrate surfaces. Solar cells, outdoor items, and packaging materials (Saber Ibrahim, 2019). One of a novel application for super hydrophobic coating, using as modified paper surface with a probability to print. In addition, Ecofriendly water-based coating was prepared in aqueous medium which safe environment from organic solvent hazards (Denisa Steinerová, 2020).

**Packaging material**

Packaging is one of the areas that attracted the attention of international organizations because of its advantages in this field that add value to the product and among these features is the protection feature (Korode la Caba, 2019). Food-packaging materials that have direct contact with food, under normal conditions of use small amounts of the material would be expected to migrate into the food, therefore, the idea of research came in an attempt to find a solution to reduce the migration of printing inks into the material (M. J. Martínez-Bueno, 2019).

## 2. Experimental

### 2.1. Materials

Styrene (St) and hydroxy methacrylate (HMA) were purified before used. Potassium peroxide (KPR), sodium dodecyl sulfate (SDS) and calcium bicarbonate were used as received. Nanosilica was prepared according to S. Ibrahim et al., 2017.

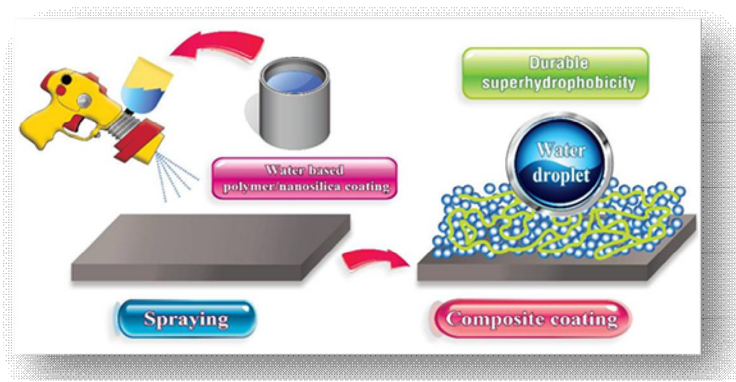


**Figure (1) preparation of super hydrophobic coating on paper**

### 2.2. Superhydrophobic (SH) coating

In 250 ml three-necked round bottom reactor, 100 ml of Millipore water was combined with 0.12 g SDS. The solution was mixed for 10minutes. 18 ml of St and HMA with stoichiometric ratio was added, and the solution was stirred for more 15 minutes at ambient temperature. After that, the temperature was raised to 70°C and KPR was added (0.17 mmol) with 320 mg Nano silicate under stirring at 600 RPM. The reaction mixture kept under heating and stirring for 4 hours under reflux. The nano superhydrophobic nano coating was applied as emulsion solution.

**Paper Coating**

Spray gun on duplex paper was applied as shown Figure 1

**Test description:**

A4 duplex paperboard was fixed on working area of spray gun in fuming hued. The coating was applied by hand spraying with working viscosity. The paper sheets are drying in thermo oven at 40 $^{o}$C for 2 hrs.

### 2.3. Test Particle size and zeta potential

The particle size distribution and zeta potential of the prepared samples were measured, using NicompTM 380 ZLS size analyzer, USA.

**Test description**

In this test, a diluted concentration of the prepared coating is used and then exposed to ultrasound (laser wavelength 750 nm) to ensure light scattering, as the beam is divided into two parts, one of which is the reference beam and the other falls on the sample, the degree of light scattering is calculated and compared with the beam Reference

**Zeta potential**

**Test description**

The purpose of this test is to measure the degree of stability and homogeneity of the emulsion in the solution. The test is carried out by applying an electric field of 4 mille volts to induce the particles to move between two electrodes and according to the speed of the particle movement; the amount of charge is calculated

### 2.4. Optical polarized microscope for ink penetration

The composite texture of coated and uncoated printed sheets was investigated using Leica DM750P polarizing optical microscope (Leica Microsystem GmbH, Switzerland). The crossed polarizers plus 530nm filters were employed for measuring the penetration depth of ink in paperboard cross section. All measurement used Leica 20X long working distance lens.

### 2.5. roughness sheets testing

The roughness of uncoated, coated and printing paperboard was measured according to RH-BR10K Paper Bekk.

### 2.6. Printing properties

1. Heidelberg offset machine SM52 MODEL 2003
2. X-Rite exact ™ Spectrophotometer

## 3. Results and discussion

### 3.1. Particle size

Particle size is one of highly interested technique to investigate the nanomaterials from the size and distribution in aqueous medium. As shown in Figure 2, the particle size of SH coating dispersed in water as eco-friendly coating is 112 nm with polydispersity index 0.03 which indicated to homogenies distribution of nanomaterials.

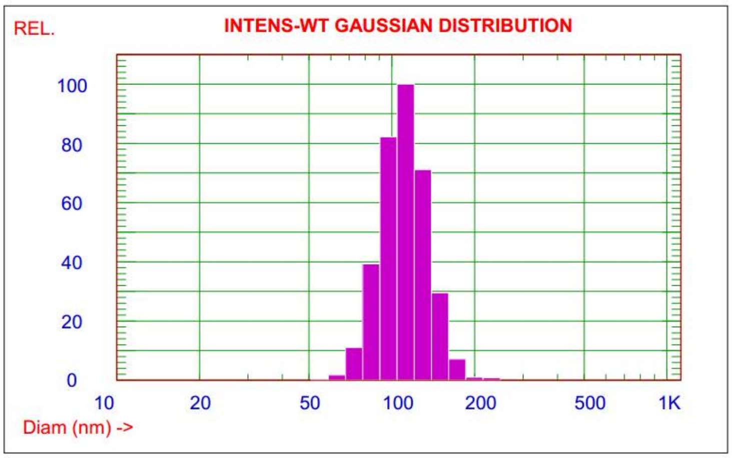


Figure (2). illustrated the particle size distribution of polystyrene/hydroxy methacrylate copolymer/SiO2 nano Coating

### 3.2. Zeta potential

Zeta potential of emulsified nanomaterial's in aqueous medium was indicated to the degree of stability against the gravity. The average zeta potential of SH coating is -43 mV which pointed to good stability of emulsion and confirm the homogenous distribution of particle as seen in particle size results. Figure 3

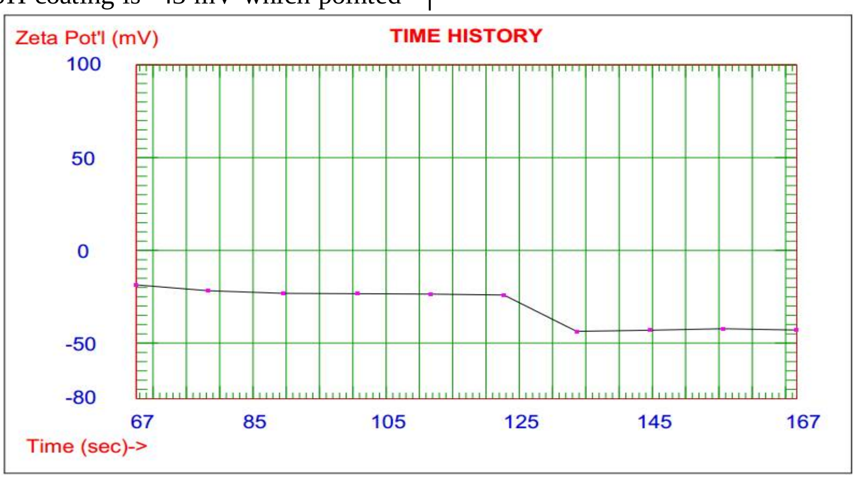


Figure 3. shows the zeta potential of polystyrene/hydroxy methacrylate copolymer/SiO2 nano Coating

### 3.3. Ink penetration

| coated | uncoated |
| --- | --- |

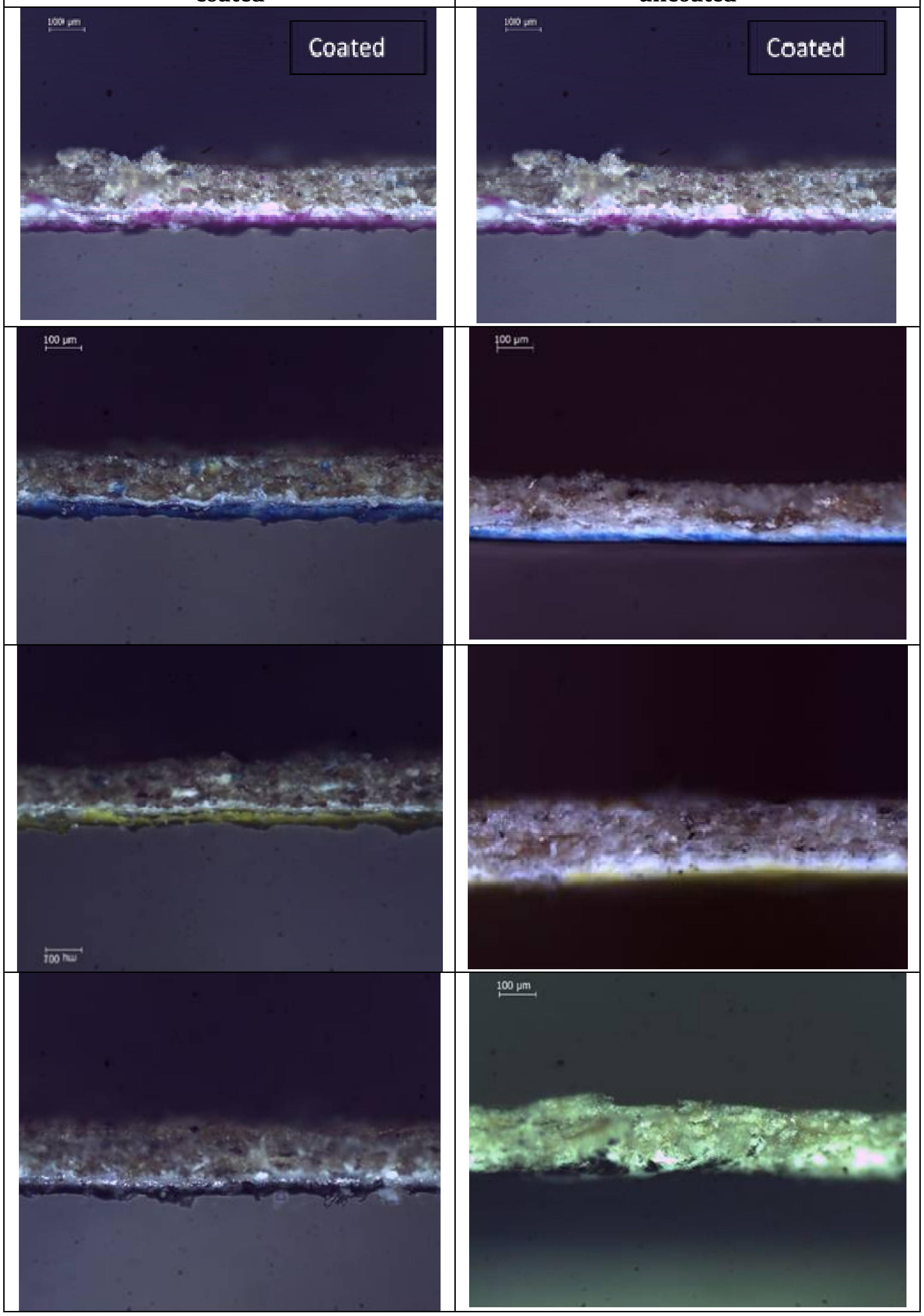


Figure 4: illustrated POM images for printing inks (black, blue, red and yellow) penetration to coated and uncoated paperboard

The previous images illustrated that, the printing ink was penetrated the surface of the printed material (duplex). Approximately 90% in the paper, either in the coated paper which penetrated at a similar rate but penetrated the SH only almost without the paper material.

If an attempt is made to dilute the print ink with ethyl acetate to bring the ink to a liquid state. simulating liquid inks was commonly used in many printing techniques. Simulating the coated and printed films are normally exposure to water (humidity and/or freezing storage), the results appear as follows in the following images.

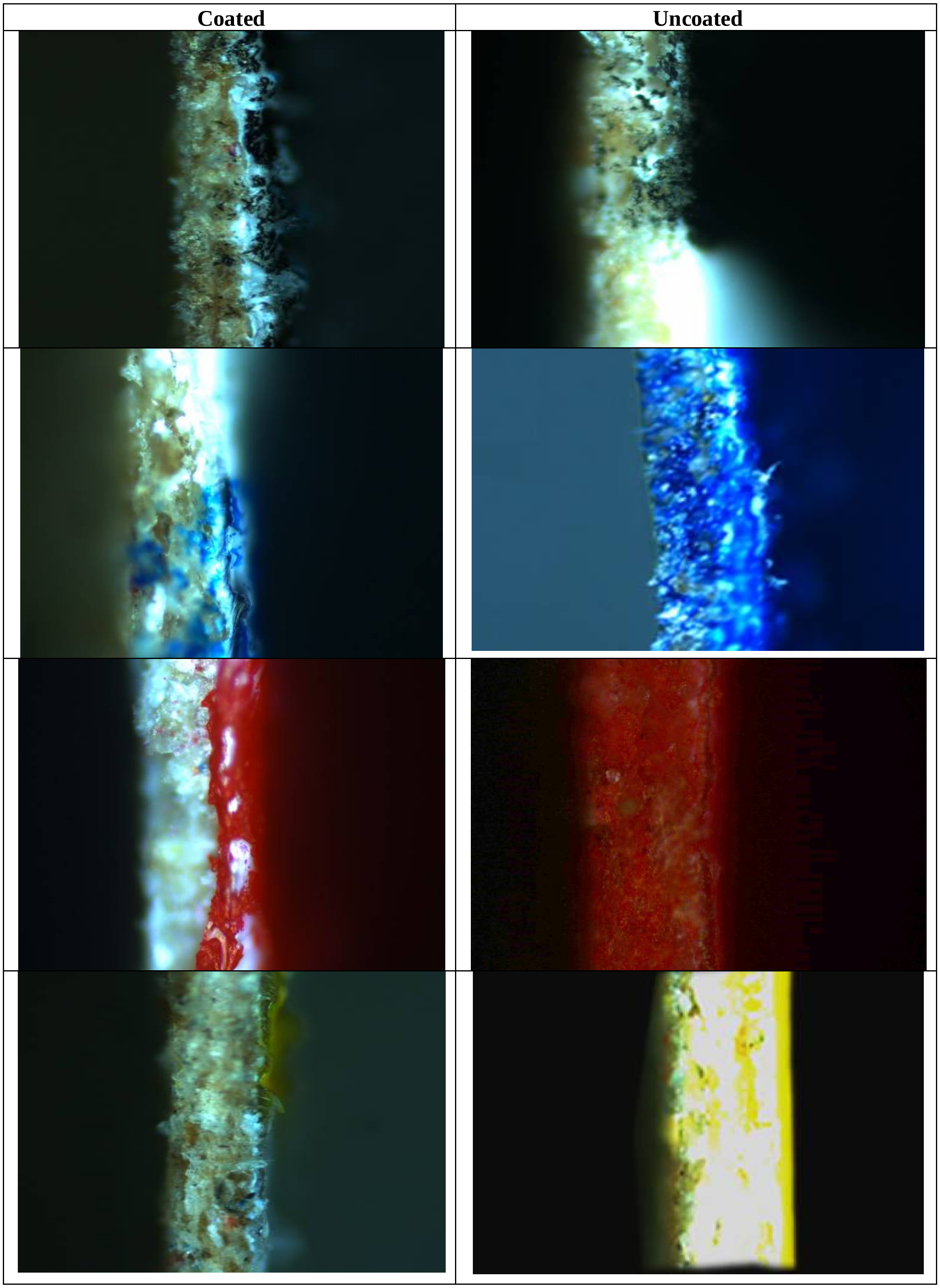


**Figure 5. illustrated POM images for printing inks (black, blue, red and yellow) penetration to coated and uncoated paperboard**

From the previous images, the penetration of the ink in the materials substrates for uncoated paper was ≈100%, while in the coated paper, with coated (SH) was a barrier to the penetration of the ink was almost more than 50%.

### 3.4. Roughness and Water Absorption

One of SH advantages is cleaning itself from any traces of dust and dirt falling on it from the atmosphere of the printing room, which is reflected in the print quality. The roughness was increased with double value for coated (4.3 µm) paper than uncoated one (2.4 µm). In addition, the printing sheet (4.9 µm) was exhibited increasing in surface roughness with 10% than coated sheet Figure 6. In addition, the water absorption of coated and uncoated paper was examined according to ISO 4046-5, 2016. The water absorption of uncoated and coated papers is 38 and 28, respectively. The degree of reduction in water uptake is 26.3 % in relative to the uncoated paper. This is pointed to the positive modification of coating on the degree of water absorption.

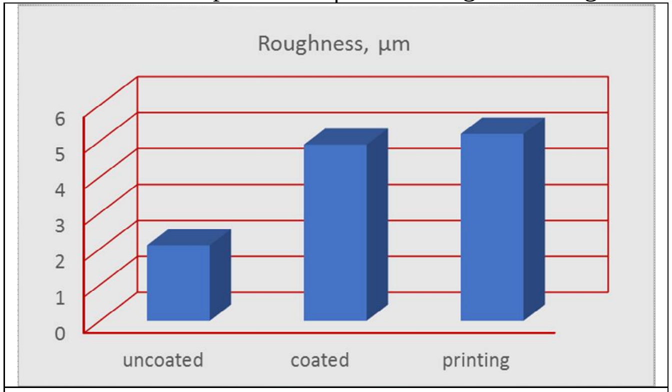


Figure 6. shows the roughness of printing, coated and uncoated paperboard

## 3.5. Printing properties

The applied study depends on making several colors measurement for the group of printing experiments to find out the effect of print ink on super hydrophobic coating and uncoated. Duplex paperboard was used as one of the most popular packaging materials on the market. Duplex paperboard, weighed 180 g/m$^2$, was divided into two parts, one group was covered with polymer super hydrophobic coting, and the other group was uncoated. Both were printed by the Heidelberg offset machine SM52 MODEL 2003, According to the following design:

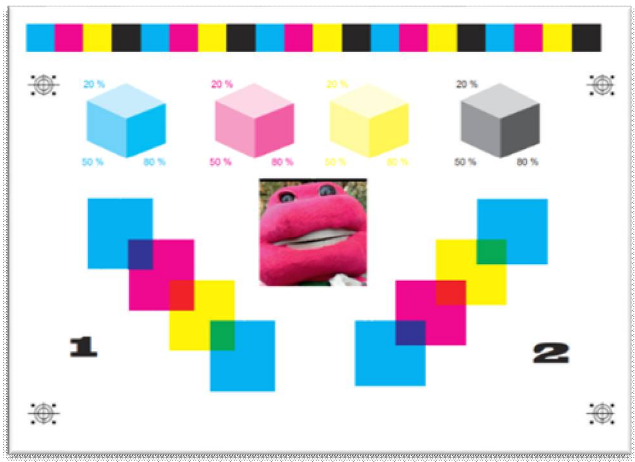


Figure 7. Shows the Printed design before and after coated

**Following display the obtained results:**

| paper comparison coated and uncoated | | | |
|---|---|---|---|
| | REF# | TEST | ΔE |
| L | 90.92 | 84.74 | **Δ3.97** |
| A | -0.16 | 0.05 | |
| B | -1.92 | -1.73 | |

| DOT GAIN FOR C | | |
|---|---|---|
| | REF# | TEST |
| 20% | 26.6 | 37.4 |
| 50% | 62.3 | 66.3 |
| 80% | 91.8 | 90.2 |

| COLOR C COMPARISON | | | |
|---|---|---|---|
| | REF# | TEST | ΔE |
| L | 54.68 | 53.09 | **Δ1.79** |

| DOT GAIN FOR M | | |
|---|---|---|
| | REF# | TEST |
| 20% | 27.6 | 40 |

| | | | |
|---|---|---|---|
| A | -33.72 | -32.22 | |
| B | -47.06 | -44.1 | |

| COLOR M COMPARISON | | | |
|---|---|---|---|
| | REF# | TEST | ΔE |
| L | 50.82 | 49.79 | **Δ1.82** |
| A | 52.01 | 60.15 | |
| B | -8.76 | -11.11 | |

| COLOR Y COMPARISON | | | |
|---|---|---|---|
| | REF# | TEST | ΔE |
| L | 85.29 | 82.23 | **Δ4.16** |
| A | -7.89 | -8.48 | |
| B | 85.49 | 69.95 | |

| COLOR K COMPARISON | | | |
|---|---|---|---|
| | REF# | TEST | ΔE |
| L | 20.93 | 23.82 | **Δ2.20** |
| A | 0.43 | 0.65 | |
| B | 2.48 | 3.33 | |

| | | |
|---|---|---|
| 50% | 64.1 | 70.3 |
| 80% | 89.5 | 89.4 |

| DOT GAIN FOR Y | | |
|---|---|---|
| | REF# | TEST |
| 20% | 30.4 | 37.8 |
| 50% | 71.9 | 68 |
| 80% | 95.6 | 91.4 |

| DOT GAIN FOR K | | |
|---|---|---|
| | REF# | TEST |
| 20% | 24 | 43.7 |
| 50% | 61 | 71.9 |
| 80% | 87.9 | 89.9 |

| DINCITY | | |
|---|---|---|
| | REF# | TEST |
| C | 1.15 | 1.02 |
| M | 1 | 0.95 |
| Y | 1.23 | 0.92 |
| K | 1.38 | 1.31 |

From the tables it can be seen coated (SH) has a great effect on dot gain, especially in the highlight areas, whether for yellow, cyan, magenta and black colors. The dot gain growth in the coated paper is greater than the dot gain growth in the uncoated paper, because the roughness of coated papers was higher than uncoated one.

Results of comparison coating (SH) with Media standard print 2016 CIE L,a,b color values of the solid base color for sheeted, web and continuous offset on eight paper grades (ISO 12647, 2016)

| COLOR C COMPARISON | | | |
|---|---|---|---|
| | REF- ISO 12647 | Test | **ΔE** |
| **L** | **56** | **53.09** | **Δ3.8** |
| **A** | **-35** | **-32.22** | |
| **B** | **-53** | **-44.1** | |

| COLOR M COMPARISON | | | |
|---|---|---|---|
| | REF- ISO 12647 | Test | **ΔE** |
| **L** | **48** | **49.79** | **Δ5.0** |
| **A** | **75** | **60.15** | |
| **B** | **-5** | **-11.11** | |

| COLOR Y COMPARISON | | | |
|---|---|---|---|
| | REF- ISO 12647 | Test | **ΔE** |
| **L** | **89** | **82.23** | **Δ7.2** |
| **A** | **-4** | **-8.48** | |
| **B** | **92** | **69.95** | |

| COLOR K COMPARISON | | | |
|---|---|---|---|
| | REF- ISO 12647 | Test | **ΔE** |
| **L** | **16** | **23.82** | **Δ6.2** |
| **A** | **0** | **0.65** | |
| **B** | **0** | **3.33** | |

The rate of color change (ΔE) between the coated and uncoated samples after making the measurements within the permissible limits according to ISO 12647. The results shown that, the colors are (cyan Δ1.79), (yellow Δ4.16), (magenta Δ1.82) and (black Δ2.20) colors.

## Conclusion

Ecofriendly Nano composites polymer coating was prepared in aqueous medium based on silicon dioxide. The superhydrophobic coating was applied on duplex paperboard through spray coating and successfully printed by litho-offset techniques. the prepared coating exhibit Nano size particle diameter with very good stability which pointed by zeta potential measurement. In addition, the water absorption was reduced after coating compared with uncoated duplex paperboard. The roughness of the coated paperboard has great value than uncoated one. Moreover, the printing properties shown that, the dot gain growth in the coated paper is greater than the dot gain growth in the uncoated paper, because the roughness of coated papers was higher than uncoated one.

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## 5. Recommendation:

1- Use an anliox cylinder to cover the coat (SH) to reduce roughness and obtain a smooth surface, thus reducing dot gain in the high-light areas.
2- The use of (SH), environmentally friend, in the field of food packaging, especially dry foods and bakery packaging.
3- Using coat (SH) in the packaging, as it stands as a barrier to the penetration of ink inside the package

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