# Polymer coating and Lamination on cellulose film

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#### SUMMARY

Due to high surface energy of Cellulose film. adhesion of plastic polymers having lower surface energy is a problem. Various pretreatment of Cellulose surface has been described which improved the adhesion both in solvent coating and lamination with plastic polymers. Extent of Heat seal strength moisture proofness, blocking, specular glass and gas barrier of various coating on Cellophane has been evaluated. The causes of loss of heat seal with time have been discussed. The technique of Super Coatings on Cellophane by PVDC disersion have been presented. The future of polymer coated Cellophane as packaging material has been discussed.

#### INTRODUCTION

Regenerated Cellulose film made a revolution in through packaging during early fifties due to its high degree of transparency and toughness. Two kinds of regenerated Cellulose film were available in advanced countries, a) Cellophane film or transparent paper (TP), and b) Ceglin film. Both films were obtained from wood cellulose, former one through Viscose process, latter one by reaction of alkali cellulose with ethylyne oxide. (Figure 1)<sup>1</sup>,<sup>2</sup>.

Cellulose film as is, has good degree of gas barrier, in fact Cellophane mask was useful as protection during second World war against chlorine gas. But there was no moisture barrier or oxygen barrier properties, which are essential for wrapping food, silverware, cigarette or tea boxes, etc. Also to make tight wrapping, sealing of end is necessary, but cellophane won't heat seal, it has to be sealed by applying adhesive solution at the seal points which is slow and can not be done in fast automatic machine. Therefore necessity for coating or lamination of Cellophane with thermoplastic polymers arose for achieving moisture proofness, heat sealability, to obtain higher oxygen barrier, etc.

In this paper, the technical and scientific pro-

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blems which were faced during development of various kinds of coating on Cellophane have been presented and the future status of such polymer coated Cellulose film in advanced countries and in developing countries has been discussed.

## EXPERIMENTAL

#### Materials

For laboratory scale anchoring and coating experiments following materials were used.

Gel film from the Plant, glycerol, Accobond 3524 (MF resin, 30%), lactic acid, hand operated wringer, wooden drying frames  $(15'' \times 12'')$ , forced draft oven, an aluminium stack of 4 frames which can be placed in the oven, etc.

For coating following resins were used :

- 1) Nitrocellulose, RS type 1/2 sec, 5/6 sec (Hercules)
- 2) Saran F 216, F 242 L (Dow Chemical)
- 3) Vinyl, VMCH and VYHH (Union Carbide)

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For lamination a small laboratory unit was used which had Corona discharge chamber in the plastic film line.

# Methods

#### a) Plasticizing and Anchoring

Gel film from plant was cut into  $15'' \times 12''$  size, was plasticised and anchored in the same bath using 5% glycerine and 0.3% Accobond at 50°C for 5 minutes Few drops of lactic acid was added to the bath to maintain the pH 4.5. The film was removed, sandwitched between two paper towels and passed through a wringer to remove excess water. The film was placed in the wooden frame so that machine direction was length wise, allowed to air drv for one hour and then placed in the oven at 90°C for 5 min. for final drying and curing. After the cure, the film was allowed to condition at 50% R. H., 75°F for 24 hours before coating.

# (a) Solvent Coating Method

Anchored film was coated with the lacquer solution on an aluminium coating frame, specially designed for the purposes (Figure 2). The conditoned film was placed in the coating frame, the frame



was clamped and exposed in the oven at 90°C for one minute before coating. Lacquer of solid content 6.8% warmed to 40°C was poured over the film and drained off one corner of the frame. The frame was placed in the oven at 90°C for one min. to remove solvents. The other side of the film was coated in the same manner and dried. Four such films  $10'' \times 6\frac{1}{2}''$  size were necessary to carry out all the tests namely, WVTR, heat seal, blocking and coating gram weight, etc.

#### (c) Lacquer Preparation

Nitrocellulose lacquer was prepared as per recommendation of Hercules Powder Company<sup>3</sup>. For Saran the resin was first wetted by toluene, allowed to soak for 15 min. at 40-45°C. then THF was added followed by other ingredients.

For Vinyl coating, VMCH and VYHH resins were first wetted with toluene, ethyl acetate was then added, mixed well and heated. The solution should be made free of gel. The other ingredients were then added and mixed thoroughly in lightninmixer which produced a clear, homegeneous solution.

# (d) Physical Tests

ASTM Test methods were followed for determining WVTR (ASTEM 96, Procedure E), Oxygen Permiability (ASTMD 1434), Haze (ASTMD 1003), Tensile (ASTMD 882). For heat sealing, sentinal heat scaler was used having flat bar seal at 275°F, dwelling time 1/2 sec. at 20 psi. For heat seal measurement suter tester was used to pull the strip and value was expressed in grams/1" width.

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# **RESULTS AND DISCUSSION**

# I Surface Preparation

Cellophane paper or any other Cellulose film is not heat sealable and has no moisture barrier properties. In order to attain above properties a thin coating of a thermoplastic polymers has to be applied on Cellophane surface. But the two surfaces, namely Cellophane and thin plastic film, are not alike. Due to the presence of large numbr of hydroxyl groups in Cellulose macromolecule, Cellulose film has high surface energy, 70-75 dynes/Cm<sup>2</sup> as compared to plastic films (30-50 dynes/Cm<sup>2</sup>) as found by zisman's contact angle measurement technique<sup>4</sup> (Table I). On the basis of fundamental theory of adhesion, it is apparent that in order to have Vander Walls force of attraction operative and also for good hydrogen bonding, the two surfaces must come into very close contact, which is not possible when the suface energies of two surfaces are not close. If they are far apart, very little contact will be established. A thin coating of a plastic film may be applied to Cellulose surface but adhesion, as measured by dry heat seal or wet anchorage test, will be very poor and such coating is not acce-ptable for partical use. But if the surface energy of Cellulose fibre could be lowered, brought down closer to plastic film by giving a surface treatment, called anchoring treatment then the adhesion improves considerably (Table II). On the other hand surface energy of plastic films also could be raised to some extent by Corona discharge treatment (TABLE I) which make them more compatiable to

# TABLE-1

# SURFACE ENERGY OF CELLOPHANE AND PLASTIC POLYMERIC FILMS

Substrates	Surface Energy	Surface after treatment
Cellophane (P1)	70-75	50-54 (anchoring)
Nitrocellulose (NC)	50-55	
Saran (F216)	40	50-55 (Corono discharge)
Vinyl (VMCH), VYHH)	42	50-55 ( do )
Polyethylene	38	48-50 ( do )
Polypro <sub>f</sub> ylene	36	48-50 ( do )

# TABLE-II

#### Pretreatment Substrate Adhesion Dry Wet Cellophane Pl None Cell/NC Poor None Cellophane P1 Cell/NC Good Fair Anchoring None Cell/Saran Poor Poor do Cell/Saran Excellent Good Anchoring do Cell/Vinyl None Poor None do Cell/Vinyl Good Fair Anchoring do Cell/PE Poor None None do " Cell/PE Good Fair Anchoring do

EFFECT OF ANCHORING TREATMENT ON ADHESION

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Cellulose surface and help the adhesion, a technique which is utilised in lamination process specially in Cellulose to polyethylene or polypropylene lamination system.

The anchoring treatment to Cellophane film containing 10-15% glycerine consists of treatment with UF, MF or PEI resin solution very small quantity (0.3-0.5% on Cellulose) followed by curing in the oven. Thus a precoat of a thermosetting resin is obtained, which lowers the surface energy of Cellophane and such film then gives good adhesion with thermoplastic resins. This technology is adopted to produce moisture proof and heat sealable polymer coated Cellophane, by all Cellophane manufactures in the World.

Thus after the surface preparation, the next is coating. The anchored dry Pl film should be coated within 4l hours. Otherwise the film develops blocking. Quality control for anchorage is done by nitrogen analysis and contact angle measurement with drops of water.

## II Nitrocellulose Coating

This is the first coating for Cellophane developed during forties much credit for which should be given to Hercules Powder Company, U.S.A. Nitrocellulose itself is an excellent film former, clear sparkling type, but it does not have any barrier properties. But it is blendable with varieties of chemicals like resin, plasticizer, clay, wax etc. which has enabled chemists to formulate with various ingredients to produce a NC lacquer which will give heat seal and moisture proofness. A typical formulation of NC lacquer has been presented in Table III.

TABLE--111 N C LACQUER FORMULATION

Ingredients	Parts by Weight	Solvents
N C 1/2 sec.	33.0	Toluene
N C 5/6 sec.	16.5	Butanol
Citroflex A-4	15.0	Ethanol
DCHP	22.5	Ethyl Acetate
Petrex T7HT	10.0	Butyl Acetate
Sunoco Wax 41	2 4.5	2
Chem 120	1.0	
Leursol 7	3.0	
Flat E 450 (34%	() 0.2	
DBP	0.2	
Citroflex A-2	5.5	

To prepare a good NC lacquer is a tricky job. We have to select resin which will give heat seal, hardness and gloss, select plasticizers which will make film flexible, softer and will give good heat

seal, Wax which will be blended with the system to give moisture proofness, we have to use fine clay to eliminate blocking etc. For dissolving we have to use both good and bad solvent, otherwise right viscosity and flow properties will not be attained.

In formulation, medium hard resin Petrex 17 HT was found to be very suitable because it gives good gloss (Figures 3A), least after yellowing (Figure 3B), and good heat sealing with DCHP plasticizer (Table IV). Wax is needed for moisture proofness but too much is not good (Figure 3C). Moisture -



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proofness as determined by standard WVTR measurement can be varied with NC coating, so also heat seal, by careful formulation of the lacquer. Thus we may have heat sealing type, MS film, nonheat sealing type M film, high WVTR, low WVTR as required by the packaging industry, (Table V). The heat seal of MS film is 150-300 gms. which is good enough for many purpose. Because of diversification of properties which can be tailor made, is possible only with NC coating and this made NC coating so popular on Cellophane paper.

# **III Saran and Vinyl Resin Coating**

Further sophistication in packaging needs led to development of Saran Coating. Saran Coating on Cellophane has excellent oxygen barrier properties besides high moisture proofness, offers protection from UV radiation of sun light, gives very high heat seal and very good wet anchorage, even under boiling water. These properties have led to wide pread use of Saran Coated Cellophane in special type of food wrapping in advanced countries.

A typical formulation of Saran lacquer is presented in Table VI. Saran formulation is very simple. The seal strength normally is 400-500 gms., which is much higher than NC coating and boiling water anchorage with 30 gms load is 50-60 seconds as compared to only 5-10 with NC Coated film. On using PEI-HCHO system of anchorage instead of Accobond (MF system) both dry and wet heat sealed could be improved to a very high level<sup>5</sup>. Such anchorge gave high gloss and slip to the film, PEI alone gives fair anchorage but it creates blocking and after yellowing. PEI-HCHO system showed improvement also for Vinyl Coating anchorage (Table VII).

Loss in strength of heat seal with Saran Coating may develop if the sealing temperature is not right and anchoring treatment is not enough. PEI-

		*R	S type	-	·			. 1	**S S ty	pe	
• •	I	Formu	la			. •		For	nula		
	1	2	3	4.		5 A.	5	6	7	8	
NC(1/2 Sec.)	50	50	50	50			50	50	50	50	parts
Resin Petrex						 1					
T7HT	36_			<u></u>		 					_ parts
Plasticizer							. *		t l		
DOP	14						14				do
DBP		14						14		,	do
DCHP	• • •		14						14		do
ТСР				14			•			14	do
Heat Seal	No	No	Slight	Slight			Slight	good	Very good	good	

# TABLE-IV

N C AND PLASTICIZER TYPE

\*R S, higher nitrogen (Will give non heat seal type coating)

\*\*S S, lower nitrogen (heat seal type coating)

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Film Code	Mois	t Proofness	Haze	Heat Seal	Tensile psi. 45%RH	Application
M S 100	High,	WVTR 0.45	3.5	175-200Ø	MD 17000 TD 8000	Bags and Ponches Fast sealing
M'S 400	High,	WVTR 0.45	do	do	MD 18000 TD 9000	Over warps, fast sealing
M S Cellophan	ne Low,	WVTR 30-60	3.0	150-175	MD 18000 TD 9000	Packing fresh Vegetables Salads etc.
MSBO film (One side coat heat sealed)	Low, ed	do	3.0	200-(Coated to Coated side)	MD 17000 TD 8000	Specially designed for lamination with PE on Un-coated side
M B O film (Non-heat sealable)	Good,	1-3	3.0	None	MD 18000 TD 9000	Monheat seal type, specially designed, for lamination, poly extrusion on Un- Coated side

# TABLE—V NITROCELLULOSE COATED FILMS

# TABLE-VI

# SARAN AND VINYL FORMULATION

Saran	*Resin F216	100 parts
	Carnaba Wax	2.5
	Stearic acid	1.25
	Clay E 999	1.20
	THF	1890
	Toluene	210

\*Copolymer of Vinyledene dichloride and Acrylonitrile

Vinyl	*VMCH	37.3 parts
	VYHH	37.3
	Polypale	3.7
	Suico Wax	5.7
	Citroflex A-4	4.4
	Clay E 999	1.4
	Chin 120	0.7
	Ethyl Acetate	1060
	Toluene	1100

\*Copolymer of Vinyl Chloride and Acrylonitrile

HCHO system of anchoring gives better retention of heat seal after storage (Table VIII).

Recent development of coating Cellophane with PVDC acqueous dispersion has eliminated use of flamable solvents like THF. Moreover WVTR is lower than solvent coating, heat seal is same, slip is good and above all coating cost has been reduced appreciably. Based on PVDC dispersion coating, British Cellophane has developed a laminated composition called Briphane which has the lowest WV TR in the film world and they claim such super coating will keep Cellophane ahead for years<sup>6</sup>.

Vinyl Coating has heat seal between Saran and NC Coating. It has good clarity because of sharp increase in price of NC cotton. Vinyl coating is becoming popular in many countries, its typical formulation has been given in a table.

# IV Lamination and Extrusion Coated on Cellophane

Cellophane film had been found to be very much suitable for both adhesive lamination and extrusion coating lamination<sup>7</sup>. Both system gives excellent barrier properties and such products are finding now wider use than 100% Cellophane. In adhesive lamination, there is no need of edge trimming, gauge control of finished product is better and slip property of PE film (low slip or high

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Coating	Type Bath 1	Bath 2	Bath 3	Heat Seal gms/2	Boiling Water test (Sec.)
Saaran	0.3% Accobond (MF)+Lactic acid	Water	9% glycerin	180	19
Saran	0.25% PE1+ Lactic acid	Water	đo	220	40
Saran	0.1% HCHO	do	9% glycerin +0.25% PEL +Lactic acid	880	400
Vinyl	0.3% Accobond +Lactic acid	Water	9% glycerin	400	20
do	0.25% PD1+ Latic acid	do	9% glycerin +0.1 HCHO	773	400
do	water	do	9% glycerin +0.25% PE1+ Lactic acid	185	37

# TABLE-VII

SUPER ANCHORING SYSTEM : FOR SARAN AND VINYL COATING

# U. S. Patent 3, 507; 685 (1970)

#### TABLE-VIII

# LOSS OF SEAL STRENGTH WITH SARAN AND P E COATING

Seal type	Sealing temp	Heat Seal after hr. Two weeks		
Sear type	1			
Saran to Saran	150	11000	6760	
do	160	10000	6280	
do	170	10000	8000	
do	180	9680	10500	
do	190	4550	8400	
Saran to Cellophan (Accobond)	e 180	400	214	
Saran to Cellophan (PEI = HCHO)	e 180	800	750	
Saran to Cellophan (No anchoring agen	e 180 t)	30 <b>0</b>	10	
PE to Cellophane (Accobond)	150	300	76	
PE to Cellophane (PE I)	150	1050	980	

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slip) does not interfere which it does in extrusion coating. On the other hand in extrusion coating there is some advantage like less Warehouse space due to resin purchase, size adjustment is easier and clarity is better. In both process, some oxidation of polymer is a requirement for good adhesion. In adhesive lamination, Corona discharge technique helps adhesicn, in extrusion process, high temperature used (500-600°F) helps to oxidise PE or PP which improves the adhesion, Although adhesive lamination has some processing advantages but extrusion coating or lamination represents the cconomically attractive way to combine two materials, Cellophanes and PE or PP.

Some examples of laminated Cellophane films which are commercially available in advanced countries have been presented in Table IX. Such films possess superior moisture proofness, high gas barrier, high durability properties, still retaining the biodegradability and good machinability character of Cellophane.

# V Future Prospect of Polymer Coated Cellophane and Conclusions

From the point of view of printability, static free property and superb machinability, Cellophane film products containing 5-10% plastic coating or

lamination, have no parallel, except oriented polypropylene (OPP) film which is emerging and has replaced Cellophane in some area of applications. However OPP needs coating to make it static free and heat sealable at lower temperature which adds up to the cost of such film. The unfavourable factor for Cellophane at present is its high cost of pro-duction. The prices of Wood Cellulose (Pulp) has gone up 40%, high grade wood Charcoal price went up 80% (needed for making CS<sub>2</sub>), caustic soda price is also higher. All these factors led to increase in cost of production of Cellophane to an alarming level. India is rich in Agriculture residues and wastes, therefore, Cellulose and carbon from such wastes<sup>8,9</sup>, should be cheaper than forest wood resources and will reduce the cost of production of Cellophane. Further cost reduction could be achieved with the development of new technology of solvent cost Cellulose film instead of going through costly Viscose process, which is in research and development stage now<sup>10</sup>.

But one factor is very favourable for Cellophane is its biodegradability and easy disposability properties which should place this packaging materials at high rating from environmental point of view. Experiments have shown that both uncoated and coated Cellophane films biodegrade and disappear under soil burial and septic tank conditions (Table X)<sup>11</sup>. The plastic films under same condition remain intact even after one year. Clogging of Municipal sewers is already taking place in cities due to indiscriminate disposal of plastic films by people. Things will get worse when our standard of living further goes up and with the rise of urban population.

## TABLE - IX

# LAMINATION AND EXTRUSION COATING COMPOSITES BASED ON CELLOPHANE

CELLOPHANE type	POLYMER type	Composition
Saran Coated	Saran	Saran-Saran (Laminate)
do	OPP	Opp/adhesive/Cello. (Laminate
Un-coated (anchored)	PE	Cell-PE (Laminated)
Saran Coated	PE	Cell/PE/Cell. (Extrusion Lamination)
Saran Coated	<b>PE+OPP</b>	Cell/PE/OPP (Extrusion Lamination)
Saran Coated	PE	Cell/PE (Laminated)
do	OPP, PE	OPP/PE/Cell/PE (Lamination and
		Extrusion)
Nitrocellulose Coated (One slde)	PE	Cell/PE (Extrusion Lamination
do	PE, foil	Cell/PE/foil/PE (Extrusion Lamination)
Nitrocellulose (non-heat seal)	PE .	MBO/PE (Extrusion coating)

TABLE---X

DIODECDADATION	OF	DACKACINC EUNS
BIODEGRADATION	Or	PACKAGING FILMS

Type Film	Soil Disintegration Days	Film Disintegration Septic Tank Half Life, Days
Uncoated Cellophane Nitrocellulose Coated	10-14	4
Saran Coated Cellophane	14—28 28—56·	220
Hydroxy ethyl Cellulose film Above, nitrocellulose coated Viaul Coated Cellus here	7—14 14—28	10 60
PE film	Remained intact one year, No weight loss	Remained intact one year, No weight loss
OPP film PVC film	do do	do do
Polyester film	do	do

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Plastic films originated from non-renewable resource (Petroleum oil is not biodegradable for easy disposal. On the other hand, Cellophane is made from renewable resources (plant Cellulose and Carbon) which were synthesised by nature through Solar energy. Cellophane is then made, it does its job, it is thrown out and it goes back to Nature thus maintaining Nature's balance as follows .--

 $\begin{array}{rcl} \text{Air} & \rightarrow & \text{CO}_2 + \text{H}_2\text{O} & \underset{\text{Energy}}{\text{Solar}} & \rightarrow & \text{Plant Cellulose} \\ & \uparrow & & \downarrow \\ & & \text{Biodegradation} & \longleftarrow & & - & \text{Cellophane} \end{array}$ 

Based on this information, a national policy should be formulated so as to retain Cellophane composites as number 1 in see through packaging market and to promote further R and D efforts to make this film available at a lower cost.

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