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### Preparation and performance of UV curable polyurethane coating for metal surfaces

Hemul V. Patel\*, Jignesh P. Raval, Pradip S. Patel

*Ashok & Rita Patel Institute of Integrated Study & Research in Biotechnology & Allied Science,  
New Vallabh Vidyanagar, Gujarat, India*

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#### Abstract

During the last 20 years, UV curable coating has achieved fast growth in the coating market. Growth of UV coatings have grown from their first application in furniture coatings to embrace many industries viz. electronics, printing and automotives due to its fast production rates, low temperature cure and reduction in volatile organic compounds (VOC). However, it faces problem regarding viscosity, in the conventional UV – cured coating system, the reactive diluents are used to reduce the viscosity. Here, we have used renewable resources in the preparation of polyurethane acrylate (PU) coatings to reduce VOC. A series of UV – curable Polyurethane acrylate prepolymers were synthesized by reacting Polyols from sesame oil (edible) using different ratio of Polyols, aromatic isocynate, and aliphatic isocynate, 2-Hydroxy ethyl methacrylate, (2-HEMA) and dibutyl tin dilaurate as a catalyst. UV curable polyurethane acrylate was formulated using reactive monomer as a diluent and Benzophenone as a photoinitiator. These coatings are cured by Ultra Violet radiation (Medium Pressure Mercury Lamp (200 Watt/inch) (280-360nm).The study of mechanical and chemical properties of cured resin films showed that hardness, and impact resistance increases with increase in cross link density (XLD). This could be due to increase in interaction of reactive monomers to the resin which increases number of double bonds and hence the XLD.

**Keywords:** Polyurethane, Urethane Acrylate, UV Radiation, (XLD) crosslink density.

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#### Introduction

UV curing has found an increasing number of industrial applications over the past decade due to its unique benefits, for example solvent-free formulations, high-cure speed and low-temperature processing. In addition to these benefits, two additional properties of UV-cured coatings are of increasing interest: scratch resistance and resistance against chemicals. One major field where

UV curing has been unable to compete with existing coating technologies in the area of outdoor applications [1]. UV-radiation curing is one of the most effective processes for rapidly transforming a liquid coating film into a solid film. This well proven technology offers a number of advantages making it suitable for the preparation of composite polymers [2]. This UV-radiation curing technology has become attractive especially in the paint, ink, and coating industries due to its very low consumption of energy and its minor emission of volatile organic compounds. By adjustment of the light intensity, ultra fast curing can be carried out at the desired rate at ambient temperature.

In general, the cure rates are slow due to the relatively unreactive nature of the unsaturated polymer internal double bond. In response to industrial demands for higher cure speed (enhanced productivities) acrylated oligomers were developed. Of the common unsaturation type, acrylate exhibits the most rapid UV, free-radical cure response in the following order: acrylate > methacrylate > allyl > vinyl. UV cured acrylated products, based primarily on urethane and epoxy backbones, fill a wide range of application needs [3].

In recent years, there has been a growing trend in using vegetable oils as renewable resources especially in oleo chemical productions. Several derivatives of vegetable oils are used as polymerizable monomers in a radiation curable system due to their environmentally friendly character and low cost when compared to products from petroleum. Moreover, the long fatty acid chains of vegetable oils impart desirable flexibility and toughness to some brittle resin systems such as epoxy, urethane and polyester resins [4]. Vegetable oils have a number of excellent properties which could be utilized in producing valuable polymeric materials such as epoxy, poly ester amide, alkyd and polyurethane, in addition to its many application in other areas [5-8]. Epoxidized vegetable oils, for example epoxidized palm oil and epoxidized soybean oil were utilized in UV-curable coating systems [9,10]. Vernonia oil, natural oil containing epoxide groups, was utilized as a polymerizable monomer in cationic UV-cured coatings [11].

In the present work we have attempted to utilize the sesame oil for the synthesis of polyurethane to convert oils into polyols. Polyols were prepared via alcoholysis of triglyceride oil by proprietary method. It was further reacted with toluene di-isocyanate and isophorone di-isocyanate in different ratio to develop a series of polyurethane. The synthesized polyurethanes were tested for the evaluation of their chemical, mechanical, thermal properties.

Diisocyanate Oligomer are widely used, because they are highly reactive and slightly toxic owing to low Vapour pressure at ambient temperature. Diisocyanate trimers (Wejchan-Judek et al. 2001) or Oligomer are used in the synthesis of polyurethane coatings and adhesives. These materials exhibit a high fire and a high thermal resistance. In the synthesis of polyurethanes generally the aromatic diisocyanates are used. The polyurethanes obtained with these diisocyanates are easily oxidized and this causes the colour change of the products. This oxidation is disadvantageous particularly in the case of coating. Therefore the aliphatic diisocyanates with linear or cyclic structures are used. However, the reactivity of these isocyanates in the presence of amino catalysts and stannous organic compounds is much lower in the synthesis of polyurethanes [12].

Polyurethanes are probably the most versatile class of polymers, owing to the great variety of raw materials that can be used in their formation. Because of its versatile mechanical, chemical, and thermal properties, polyurethane has found many applications in various fields. Polyurethanes are usually used as adhesives, coatings, foams, and different kinds of plastics and elastomers, as well as matrix for polymeric composites [13, 14]. The unique feature of polyurethane based composites is that they can exhibit usable ranges of deformations much larger than those of composites with stiffer matrices, such as metals, ceramics, or rigid polymers [15].

## Material and Methods

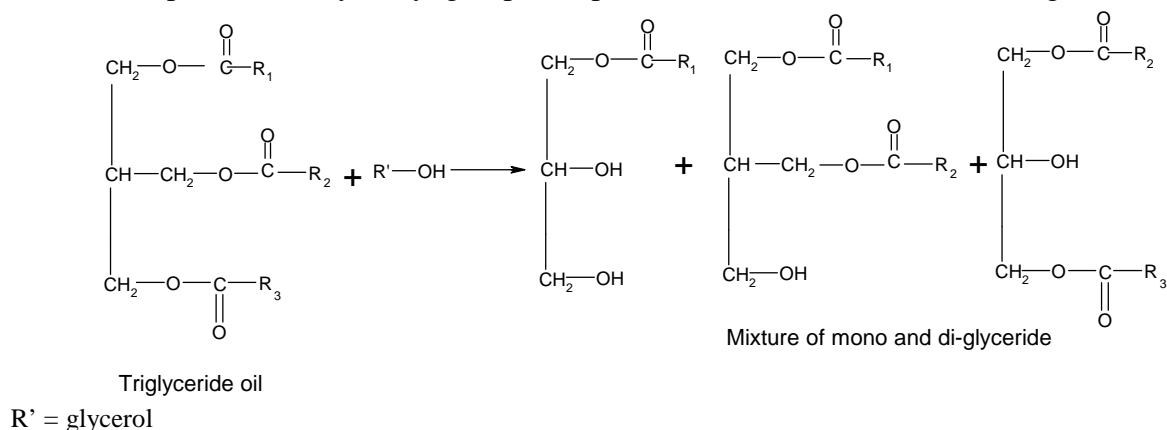
### Experimental

In the present study, alkali Refined Sesame seed oil was obtained from local market. Trimethylolpropane (TMP) was obtained from Himalaya Resins, Halol, Gujarat and Ethylene glycol and Dipropylene glycol (Merck, USA) was used as received. Aromatic and Aliphatic Isocyanates (TDI, IPDI) were supplied by Grand polycoats, Padra, Vadodara, India. Lithium hydroxide (LiOH) was procured from Astron Chemicals, Ahmedabad, India. Hydroxyethylmethacrylate (HEMA) was obtained from Himalaya Resins, Halol. Dibutyltindiluarate (DBTDL), Photoinitiator and Reactive diluents were obtained from Merck, USA.

### Resinification of the Oil

#### Preparation of monoglyceride of the oil

A three-necked round-bottomed flask, equipped with a mechanical stirrer, thermometer and nitrogen gas inlet, was used for preparation of the Sesame oil based Polyols. The reaction vessel was flushed with nitrogen and charged with sesame oil and Ethylene glycol, Di-propylene glycol and Trimethylol propane (TMP) with different ratio (1:0.75), (1:1), (1:2). and Lithium hydroxide 0.05 wt % (with respect to the oil) under continuous stirring. The mixture was then heated up to  $240 \pm 5$  °C until it formed the monoglyceride, which was confirmed by its solubility in methanol (resin: methanol = 1:3 vol/vol). The resultant polyols were dried at 80°C under vacuum and their characterization is shown in Table: 1. FTIR analysis of oil and polyols has been carried out to confirm the presence of hydroxyl group. A representative reaction is shown in Fig 1.



**Figure: 1 Preparation of monoglyceride of the oil**

**Table: - 1 Characterization of Sesame seed oil based Polyols**

Sr No.	Description Code	Polyol Type	Oil:Polyol Ratio	OH Value	Viscosity @ 30°C	Color	Specific Gravity	Iodine Value	%OH Group
1	SE-1	Ethylene Glycol	(1:0.75)	90.10	200	10	0.9265	110	3.23
2	SE-2	Ethylene Glycol	(1:1)	115.10	200	10	0.9898	109	5.95
3	SE-3	Ethylene Glycol	(1:2)	220.00	250	12	0.9958	108	6.98
4	SD-1	Dipropylene glycol	(1:0.75)	80.00	200	10	0.9708	109	2.80
5	SD-2	Dipropylene glycol	(1:1)	110.00	200	12	0.9918	106	4.85
6	SD-3	Dipropylene glycol	(1: 2)	195.22	225	12	0.9958	104	5.65
7	ST-1	Trimethylol Propane	(1:0.75)	125.0	200	10	0.9950	106	4.67
8	ST-2	Trimethylol Propane	(1:1)	160.0	250	13	0.9985	105	6.72
9	ST-3	Trimethylol Propane	(1:2)	294.21	250	13	0.9998	105	7.88

E – Ethylene Glycol, S – Sesame oil, T – Trimethylol propane

#### ***Preparation of Urethane acrylate Oligomer***

The Oligomer was prepared by two step reaction. In the First step different polyols and aromatic or aliphatic isocyanate (1:2 mole) were charged into a 250 ml four – necked round bottom flask equipped with a mechanical stirrer, thermometer, condenser with drying tube, and a nitrogen gas inlet system, approximately 0.03 wt % of DBTDL was added to the system, and the mixture heated to 55 to 60 °C and maintained for 1 hr with stirring.

**Table: 2 Sesame oil based urethane acrylate Oligomer: (aromatic) (TDI)**

Code	Mole ratio	OH value	OH Equi.wt	TDI Equi.wt	HEMA Equi.wt	% Wt Composition		
						Polyol	TDI	HEMA
SE-1ar	(1:0.75)	90.10	622.64	87.08	130	67.20	18.79	14.03
SE-2ar	(1:1)	115.10	487.40	87.08	130	61.57	22.00	16.42
SE-3ar	(1:2)	220.00	255.00	87.08	130	45.60	31.14	23.24
SD-1ar	(1:0.75)	80.00	701.25	87.08	130	69.74	17.32	12.93
SD-2ar	(1:1)	110.0	510.0	87.08	130	62.64	21.39	15.39
SD-3ar	(1:2)	195.22	287.36	87.08	130	48.57	29.44	21.97
ST-1ar	(1:0.75)	125.0	448.80	87.08	130	59.60	23.13	17.26
ST-2ar	(1:1)	160.0	350.62	87.08	130	53.54	26.59	19.85
ST-3ar	(1: 2)	294.21	190.68	87.08	130	38.53	35.19	26.27

The reaction progress was determining unreacted –NCO groups by dibutylamine back titration method<sup>[18]</sup>.

**Table: 3 Sesame oil based urethane acrylate Oligomer: (aliphatic) {IPDI}**

Code	Mole ratio	OH value	OH Equi.wt	IPDI Equi.wt	HEMA Equi.wt	% Wt Composition		
						Polyol	IPDI	HEMA
SE-1al	(1:0.75)	90.10	622.64	111.29	130	63.58	22.80	13.33
SE-2al	(1:1)	115.10	487.40	111.29	130	58.04	26.47	15.48
SE-3al	(1:2)	220.00	255.00	111.29	130	41.98	36.60	21.40
SD-1al	(1:0.75)	80.00	701.25	111.29	130	66.56	21.09	12.33
SD-2al	(1:1)	110.0	510.0	111.29	130	59.14	25.77	15.07
SD-3al	(1:2)	195.22	287.36	111.29	130	44.92	34.75	20.32
ST-1al	(1:0.75)	125.0	448.80	111.29	130	56.02	27.74	16.22
ST-2al	(1:1)	160.0	350.62	111.29	130	49.88	31.62	18.49
ST-3al	(1: 2)	294.21	190.68	111.29	130	35.11	40.93	23.94

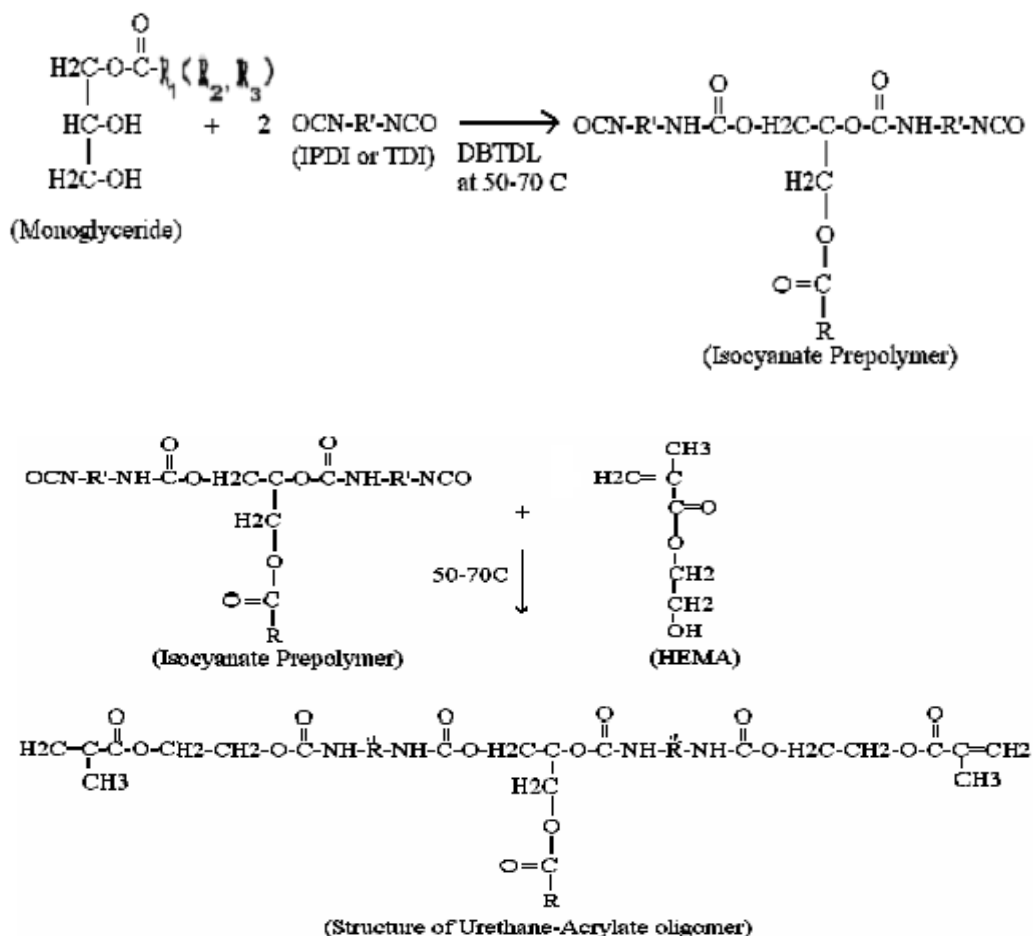
**Table: 4 Physical properties of urethane acrylate (Aromatic)**

Description Code	Colour	Sp.Gr. (wt/ltr.)	% NVM	Viscosity(cPs)
SE-1ar	15	1.02	98.85	1248
SE-2ar	14	1.03	99.50	2535
SE-3ar	12	1.05	99.87	3578
SD-1ar	13	1.02	98.31	1290
SD-2ar	13	1.02	98.99	2590
SD-3ar	10	1.04	99.87	3520
ST-1ar	14	1.05	99.65	2856
ST-2ar	13	1.05	99.69	3876
ST-3ar	10	1.06	99.78	4925

**Table: 5 Physical properties of urethane acrylate (Aliphatic)**

Description Code	Color	Sp.Gr. (wt/ltr.)	% NVM	Viscosity (cPs)
SE-1al	14	1.02	98.98	1380
SE-2al	12	1.03	99.84	2450
SE-3al	10	1.05	99.87	3320
SD-1al	14	1.04	99.76	1495
SD-2al	12	1.05	99.55	2203
SD-3al	10	1.05	99.88	3210
ST-1al	14	1.05	99.78	2750
ST-2al	13	1.06	99.97	3800
ST-3al	10	1.06	99.78	4780

In the second step, HEMA was added dropwise after the reaction mixture was cooled to 45<sup>0</sup>C, and the reaction continued for 4h. When the amount of remaining isocyanate in the system was less than 0.1 wt%, the temperature was raised to 75<sup>0</sup>C for another 0.5h to eliminate the residual NCO group. A stream of dry nitrogen gas was led into flask and maintained throughout the reaction. A representative reaction scheme is shown in Fig 2. The weight composition of the reactants used in urethane acrylate production is as shown in Table 2,3,4 and 5.



**Figure: 2 Preparation of Urethane acrylate Oligomer**

### ***Preparation of UV Curable coating composition***

The urethane acrylate oligomer was taken in a beaker and after addition of stoichiometric amount of reactive diluent and photoinitiator stirred at room temperature. The amount of each ingredient used in formulation is shown in Table 6.

### ***Application and curing of coating composition***

Sample to be tested for UV – curing was coated onto MS steel test panel (15cm X 5cm) as follows. An excess of the sample was placed at one end of the test panel and using a rod applicator (K-Bar No.5) drawn across the substrate with even pressure pushing excess material off the edge. The coated panel was exposed to a (200 Watt/inch) (280-360nm) medium pressure

mercury vapor lamp of UV-2KW-2-35 curing system. This method produced coating with average wet film thickness of 23-35  $\mu\text{m}$ .

**Table: 6 Composition of UV Curable coatings**

Sr.No	Ingredient	Wt%	Function
1	Urethane acrylate	70%	Oligomer
2	TMPTMA	25%	Reactive diluents as Crosslinking monomer
3	Benzophenone	3.50%	Photoinitiator
4	DMEA	1.50%	Activator/Catalyst
	Total	100%	

TMPTMA - Trimethylol propane trimethacrylate, DMEA - Dimethyl Ethanol amine.

#### ***Characterization of sesame seed oil based Polyols***

The sesame oil based Polyols was characterized for Hydroxyvalue (OH), Viscosity, colour, Specific gravity, Iodine value per the standard Methods [16]. The properties are reported in Table1.

#### ***Characterization of Urethane Acrylate Oligomer***

The urethane acrylate oligomer prepared as above was free flowing liquid similar to the conventional oligomer. Various characteristics of this oligomer were determined as per the standard methods [17]. The properties like non volatile content (%), colour and viscosity (Brookfield viscometer, RV-II (cPs), and density (wt/ltr) were determined. The results are reported in Table 5.

This urethane acrylate was also characterized by instrumental methods like IR- Spectroscopy (Nicolet-FTIR spectrophotometer).

#### ***Evaluation of coating***

The coatings were evaluated within one hour after irradiation. The coatings were examined for various mechanical properties like adhesion, flexibility, impact resistance and scratch hardness as per ASTM D 3359-97, ASTM D0522-93 and ASTM 2197 respectively. The impact resistance of dried film was checked by Tubular impact tester. The films were also evaluated for their chemical, corrosion and solvent resistance as per methods of their characterization described in the literature<sup>[22]</sup>. The results of the film characterization are reported in Table.7, 8.

## **Result and Discussion**

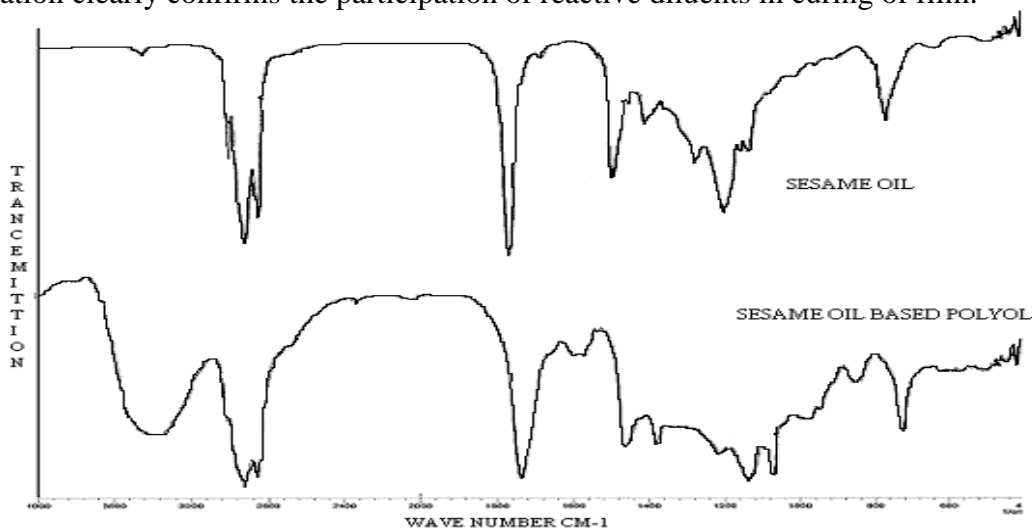
#### ***Characterization of sesame oil based polyols***

The obtained sesame oil – based polyols was a viscous yellowish liquid with viscosity of 250 poise which was much higher than that of the regular sesame oil. This high viscosity is due to hydrogen bonding associated with the hydroxyl groups. The hydroxyl values (Tab.3) clearly indicate formation of polyols. The presence of hydroxyl group in the sesame oil based polyol is also reflected by the transmittance peaks at wave numbers of  $3375\text{ cm}^{-1}$ ,  $1100\text{ cm}^{-1}$  (due to  $-\text{OH}$  secondary alcohol) (Fig.-3), and  $1050\text{ cm}^{-1}$  (due to  $-\text{OH}$  of primary alcohol). Furthermore, the obtained product was found to have complete solubility in ethanol.

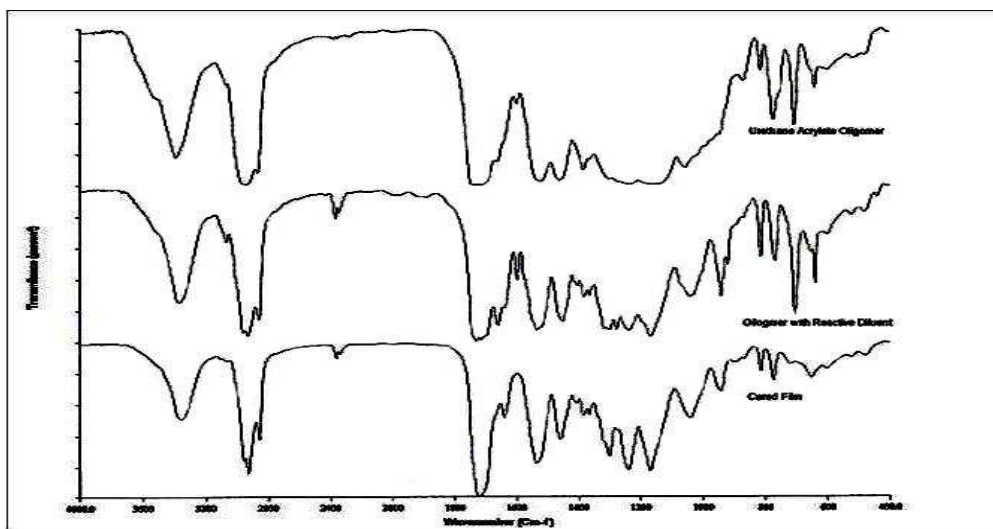


### ***Characterization of Urethane acrylate Oligomer***

The IR spectra were scanned for monitoring the presence of unsaturation in the resin (due to acrylate groups) as well for their utilization during UV-radiation giving cured coating films. The disappearance of the band at  $815\text{ cm}^{-1}$ – $770\text{ cm}^{-1}$  frequency region was monitored for the extent of curing. It was found that the band intensity increases as the urethane acrylate resin was mixed with reactive diluents (Fig.4) and after the irradiating of coating compositions with UV light the intensity decreases significantly as shown in the IR spectrum of cured film (Fig.4). This observation clearly confirms the participation of reactive diluents in curing of film.



**Fig: 3 FTIR spectra for oil and Polyol**



**Figure: 4 FTIR Spectra of Polyurethanes**

### ***Mechanical & chemical Properties***

Adhesion of the cured film was found to be better in experimental sets with oil:polyol ratio 1:1 and 1:2. Conversely the lower ratio showed poor adhesion which can be attributed to poor film



strength due to high oil proportion (SE-1ar, ST-1ar, SD-1ar and SE-1al, ST-1al, SD-1al). Also certain compositions with oil:polyol ratio 1:1 & 1:2 having TMPTMA as Crosslinking reactive diluent showed brittle failure. This could be due to very high crosslink density in the cured film (SE-3ar, ST-3ar, SD-3ar, & SE-3al, ST-3al, SD-3al). The results of flexibility and impact resistance showed the same trend as that shown above. Tables 7 and 8 shows the various chemical properties of different Polyurethanes. The result of scratch hardness of cured film was found to be very interesting and encouraging. Experimental sets (SE-3ar, ST-3ar, SD-3ar, & ST-3al, SD-3al, SE-3al) showed excellent scratch hardness. The improvement in the scratch hardness can be related to the aromatic isocyanate as a proven hard crosslinker and also the higher crosslink density (XLD) in these experimental sets.

**Table: 7 Mechanical property of UV cured coatings based on sesame oil (Aromatic)**

Description Code	Scratch Hardness (gms)	Impact Hardness (lb) inch	Pencil Hardness	Flexibility 1/8" mendrel	Cross Hatch Adhesion (%)	DFT Microns (μ)
SE-1ar	1000	F	3H	F	F	23
SE -2ar	1500	F	3H	P	P	23
SE -3ar	2100	P	4H	P	P	24
SD-1ar	900	F	3H	F	F	22
SD- 2ar	1800	P	3H	P	P	23
SD-3ar	1800	P	4H	P	P	23
ST -1ar	900	F	2H	F	P	24
ST- 2ar	1900	P	4H	P	P	26
ST -3ar	2000	P	5H	P	P	26

P-Pass F-Fail,Hardness: 6H>5H>4H>3H>2H>1H>H>HB>1HB>2HB>3HB>4HB>5HB>6HB

The solvent resistance is the inherent property of thermosetting (convertible) coatings, the degree of curing and no. of crosslinks (XLD) affects the solvent resistance to a greater extent.

**Table: 8 Mechanical property of UV cured coatings based on sesame oil (Aliphatic)**

Description Code	Scratch Hardness (gms)	Impact Hardness (lb) inch	Pencil Hardness	Flexibility 1/8" mandrel	Cross Hatch Adhesion (%)	DFT Microns (μ)
SE -1al	700	F	2H	F	F	23
SE -2al	1000	P	2H	P	P	24
SE -3al	1800	P	3H	P	P	23
SD-1al	700	F	3H	F	F	23
SD- 2al	900	P	3H	P	P	23
SD -3al	1500	P	4H	P	P	24
ST -1al	900	F	3H	F	P	23
ST- 2al	1500	P	2H	P	F	25
ST -3al	2100	P	3H	P	P	27

P-Pass F-Fail,Hardness: 6H>5H>4H>3H>2H>1H>H>HB>1HB>2HB>3HB>4HB>5HB>6HB

The results of MEK double rub test was taken as a measure of solvent resistance. Compositions SE-3ar, ST-3ar, SD-3ar, & SE-3al, ST-3al, SD-3al showed excellent MEK resistance. The chemical and corrosion resistance reveals that experimental sets with high oil content (SE-1ar, ST-1ar, SD-1ar and SE-1al, ST-1al, SD-1al) gave poor performance. This can be attributed to the hydrolysable ester linkage of triglyceride (oil) as well as lower XLD resulting into higher permeability of corrosive materials. Whereas composition SE-3ar, ST-3ar, SD-3ar, & ST-3al, SD-3al, SE-3al showed excellent chemical resistance (acid and alkali resistance) and corrosion resistance. Tables 9 and 10 shows the various chemical properties of different Polyurethanes.

**Table: 9 Chemical property of UV cured coatings based on sesame oil (Aromatic)**

Description Code	Acid resistance 5%HCl	Alkali Resistance 5%NaOH	Corrosion Resistance 5%NaCl	MEK Double rub
SE-1ar	4	4	4	55
SE -2ar	5	5	5	74
SE -3ar	5	5	5	85
SD-1ar	4	4	3	65
SD -2ar	4	5	4	75
SD-3ar	5	5	5	90
ST -1ar	4	4	5	55
ST-2ar	5	4	5	75
ST -3ar	5	5	5	95

**Table: 10 Chemical property of UV cured coatings based on sesame oil (Aliphatic)**

Description Code	Acid resistance 5%HCl	Alkali Resistance 5%NaOH	Corrosion Resistance 5%NaCl	MEK Double rub
SE -1al	3	5	4	45
SE -2al	5	3	4	65
SE -3al	5	5	5	85
SD -1al	4	3	3	65
SD- 2al	4	4	4	65
SD -3al	5	5	5	80
ST -1al	5	5	5	85
ST- 2al	5	5	5	95
ST -3al	5	5	5	95

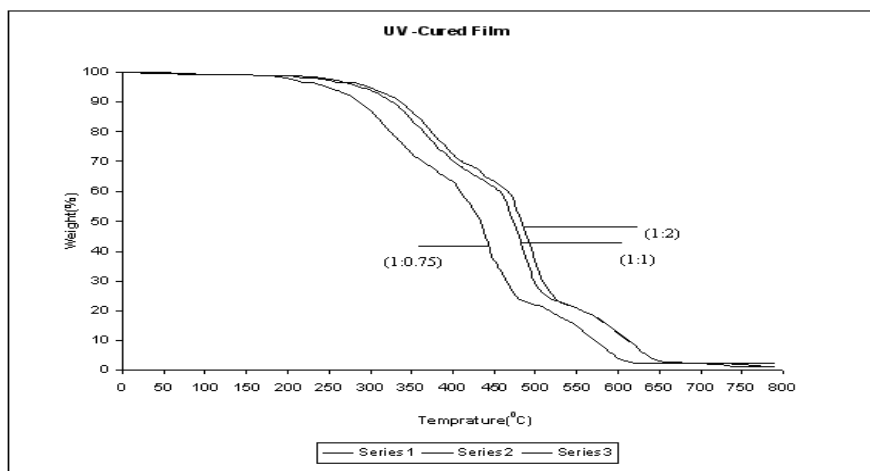
0 - Film completely removed  
2 - Film partially cracked  
4 - Slight loss of gloss

1 - Film cracked and partially removed  
3 - Loss of gloss  
5 - Film practically unaffected.

### ***Thermal Stability by TGA Analysis***

TGA thermo grams of cured films of the sesame oil derivatives are shown in Fig.5. TGA thermo gram clearly indicate good thermal stability of the cured film up to 100<sup>0</sup>C with minimum

amount of weight loss, which may be due to minimum amount of unreacted components, and TGA thermo gram also indicates that degradation occur in three steps. The initial decomposition temperatures of all the cured film are near about 200 °C. And the maximum decomposition temperature of all the cured film are above the 600 °C. This indicates that all the cured film has good thermal stability.



**Figure: 5** Thermo Gravimetric Analysis of UV-Cured films

## Conclusion

The UV curable coating compositions were successfully prepared using sesame seed oil as a renewable resource of raw material. Optimization of various parameters is done and optimum experimental sets are ST-3 al, ST-3aro, SD-3Ali, SD-3aro, SE-3 Ali, SE-3aro. Looking to the results of chemical resistance, solvent resistance and excellence mechanical properties and gloss property, the above compositions are suggested to be useful as a high performance UV-curable coating.

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## References

- [1] PS Karmee; R Mahesh, *J. of Am. Oil. Chem. Soc.*, **2004**, 81, 425.
- [2] C Decker; Kinetic Study and New Applications of UV Radiation Curing. *Macromol. Rapid. Commun.*, **2002**, 23, 1067.
- [3] US Patent , 3,907,865, **1973**.
- [4] K Prashnatha; VK Pai, BS Sherigara; S Prashanna Kumar, *Bull. Master. Sci.*, **2001**, 24, 535.
- [5] SK Das; S Lenka, *J. Appl. Polym. Sci.*, **2000**, 75, 1487.
- [6] VJ Crivello; R Ghoshai, US Patent, 5, 318, 808 (**1994**).
- [7] DS Treybig; DS Wang; PS Sheih; LA Ho, U.S. Patent, 5,151,485 (**1992**).
- [8] K Lawson, *Catching New Light under Loosened Regulations*, *Chemical Week*, **1996**.

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- [9] WD Wan Rosli; RN Kumar; S Mek Zah; M Hilmi Mohd, *Eur. Polym. J.*, **2003**, 39, 593-600.
- [10] R Raghavachar; G Sarnecki; J Baghdachi; J Massingill, *J. Coat. Tech.*, **2000**, 72,125.
- [11] SF Thames; H Yu, *Surf. Coat. Tech.*, **1999**, 115, 208.
- [12] HG Schmelzer; *Polyurethane World Congress*, **1987**, 614.
- [13] S Benli; Y Yilmazer; F Pekel; S Ozkar, *J. of Appl. Polym. Sci.*, **1998**, 68, 1057-1065.
- [14] ZA Mohd Ishak; PY Wan; PL Wong; Z Ahmad; US Ishiaku; J Karger- Kocsis, *J. of Appl. Polym. Sci.*, **2002**, 84, 2265–2276.
- [15] ZM Huang; S Ramakrishna; AA Tay; *Composites Science and Technology*, **2000**, 60, 671-691.
- [16] Gardner Sward, *Paint Testing Manual Physical and chemical Examination of Paints, Varnishes, Lacquers and Colors*. Thirteenth Edition, ASTM. **1972**.
- [17] Paul Thomas, *Waterborne & Solvent based Surface Coating Resins and their Application*, **1999**, Volume: III, 54-55.