



## Research Journal of Pharmaceutical, Biological and Chemical Sciences

### Study of copper 12-Hydroxyl oleate and copper 12- Hydroxyl oleate blended with starch in polypropylene to improve photo and biodegradation

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#### ABSTRACT

A new additive Copper (Cu) 12-hydroxyoleate was successfully synthesized and then the Cu 12-hydroxyl oleate (Cu oleate) blended with starch (Cu /S), and it's both additive the performance on the photodegradability of polypropylene (PP) films were evaluated by observing the disparity in the structural characteristic, surface morphology, mechanical, thermal properties upon degradation. The PP films blended with different compositions (1, 2 & 3 wt %) of Cu oleate and Cu /S were prepared by melt blending. The Cu oleate as well as Cu /S showed greater activity as photo initiator and as bioactive components. The photodegraded PP films were subjected to biodegradation in the presence of the microbes isolated from a dump. Fragments occur progressively in the biodegradation of the photodegraded films. Moreover, the biodegradation test results reveals that the Cu oleate and Cu /S shows 19% and 24% respectively, of biodegradation on photodegraded PP films when observed at the end of 45 days.

**Keywords:** Photodegradation; Copper12-hydroxyoleate; Copper12-hydroxyoleate/Starch; Polypropylene; Biodegradation.

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## INTRODUCTION

The great use of polymers in a lot of manufacturing and domestic fields causes environmental problems connected with their operation. Some synthetic plastics including polyolefins, as polyethylene, are characterized by relatively high stabilities under both photochemical and environmental conditions. The use of natural or synthetic photo and biodegradable polymers is promising [1]. A variety of environmental factors that may render polymer degradation are: oxygen, temperature, sunlight, water, stress, living organisms, and pollutants [2]. Many studies have been carried out by irradiating the materials in the presence or absence of oxygen, with UV-radiation having wavelength near to those of the UV components of solar light [4-8]. Low density polyethylene in the packaging field and its inherent resistance towards degradation preparation of polymer film having varying amounts of oxidized polyethylene and evaluation of its degradation behavior as a function of irradiations time [9]. The biodegradability of synthetic polymers like polyethylene can be enhanced by the addition of biodegradable additives to the formulation of plastics [10]. The biodegradability of the polyethylene-starch blends can be further enhanced by the addition of other additives such as auto oxidants (for example unsaturated fatty acids and their derivatives), photo degraders (for example aromatic or aliphatic ketone), chemical degraders (for example an aliphatic polyhydroxy carboxylic acid) and various compatibilizers [11-13]. The usage of substances that catalyze the thermal or photo-oxidative degradation, known as pro-degradant agents, which are generally composed by transition metals (zinc, copper, cobalt, Manganese etc), has been evaluated by some authors [14]. Bikiaris and collaborators [15] study the effect the use of cobalt stearate in LDPE /Starch Blends whereas Roy [16] evaluated the influence of the latter agent in the degradation of LDPE films. This photo oxidative and biodegradable additive combination increases in low molecular weight fraction by chain scission in polyethylene, thereby facilitating increasing the percentage of biodegradation. They have explained the mechanism of the degradation process and showed that polyethylene is degradable giving end products as Carbon dioxide (CO<sub>2</sub>) and water (H<sub>2</sub>O) [17]. Scientific evidence for the oxo-biodegradation of hydrocarbon polymers, Carbon-chain polymers vary remarkably in their ability to resist per oxidation [18]. Polyvinyl chloride > polyethylene > polypropylene > cis-polybutadiene > cis-polyimide. The following sequence shows some common commercial hydrocarbon polymers in order of decreasing oxidative stability. The sequence reflects the ease of hydrogen abstraction by peroxyl. Because of its ease of oxidation and loss of mechanical properties. In the present study was performed on PP (after PE) keeping in mind its prime importance in the packaging field and its inherent resistance towards degradation. This paper describes the preparation of polymer sheeting having varying amounts of Copper 12-hydroxyoleate Copper12-hydroxy oleate (oxidized polypropylene) mixed with starch and evaluation of its photo and bio degradation behaviors as a function of irradiation time.

## MATERIALS AND METHOD

Copper (II) sulphate penta hydrate, Starch (from Maize product) powder, Sodium hydroxide, Ricinoleic acid (12-hydroxy oleic acid) were used without further purification. The

MFI for the polymer was 3.4 g/10 min, with crystalline melting point of 165°C and density of 0.94 g/cm<sup>3</sup>. General purpose film grade PP H034SG has been used to prepare films. Milli Q ultrapure water was used throughout the course of this work.

### **Synthesis and Preparation of Copper 12-hydroxylolate [19]**

#### **Blending and film preparation of PP**

The Cu 12- hydroxyl oleate in different formulation such as 1%,3% and 5% and then Cu 12-hydroxyl oleate 0.5% / starch 0.5% was melt blended with PP at three different formulations 1, 2 & 3% in (Haake, Rheomex OS, PTW16, Thermo scientific, Germany) Modular Torque haake Rheometer. The PP was blended with the synthesized Cu oleate and Cu /S in varying percentages (1%, 2% and 3%), by using Torque Rheometer, Blending was carried out at a temperature range of 210, 200, 190,180 and 150°C at a screw speed of 75rpm. Subsequently, the pellets are dried in a dehumidifier at 70°C for two hours to remove moisture. The film was prepared by using film die for all the three percentages of additives. The wall thickness of the film was kept as 50 microns by controlling the speed of the nip rollers and output rate.

#### **Photodegradation**

All blended samples were subjected to photodegradation studies using QUV UV Weather-o-meter. Films of 25mm width were used to evaluate the degradation phenomenon. Samples were exposed to two different test cycles of UV irradiation and condensation as shown in Table 1. Subsequently the samples were tested and characterized for 1 and 2 (for 75 hours degrade). For 24 hours for eight hours UV cycle was carried out and for next four hours condensation cycle was carried. For the remaining 12 h, the cycle was repeated in the same order. Photodegradation affect the physical and optical properties of a plastic relative to the initial specified properties. The mechanism of Norrish type I degradation [8, 17].

#### **Fourier Transform Infrared Spectroscopy (FTIR)**

The structural changes in PP films due to the presence of Cu oleate and Cu / S effect of UV exposure were studied by Nicolet 6000 (USA) Fourier Transform Infrared Spectroscopy (FTIR) with the wave number range of 400-4000 cm<sup>-1</sup>. Evaluation of the photodegradation by ATR-IR was performed by following the increase in the carbonyl absorbance peak around 1710-1750 Cm-1 which is characteristic of LDPE (i.e.,-C-H bending modes in polyethylene) was used as a reference peak or internal standard. The results were expressed by carbonyl index defined as  $A_{1710}/A_{1450}$  [8, 14].

$$\text{Carbonyl Index (CI)} = \frac{\text{Absorption at 1710 Cm-1 (Maximum carbonyl peak)}}{\text{Absorption at 1450 cm-1(Internal standard)}}$$



## **Thermal Properties**

### **Differential Scanning Calorimeter (DSC) Analysis**

Melting behavior of Cu oleate as well as Cu /S blended polypropylene is being studied by employing Perkin Elmer (USA) differential scanning calorimeter. Sample 5 mg weight were scanned from 45 to 200°C at the heating rate of 5°C/min to detect the melting characteristics of the sample before and after exposure to UV radiation.

### **Thermo gravimetric analysis (TGA)**

Thermal degradation of PP-Cu oleate and Cu /S blended sample of before and after UV Exposure were analyzed by Perkin Elmer (USA), at the heating rate of 10°C/min from 50 to 700°C.

### **Melt flow Index (MFI)**

The MFI of the Cu oleate and Cu /S blended PP were determined before and after UV exposure according to ASTM D 1238 at 230°C under the load of 2.16 kg using Melt Flow Index tester, Loyd Instruments, UK.

### **Scanning Electron Morphology (SEM)**

The scanning electron microscopic analysis of fractured surface of PP- Cu oleate as well as Cu /S film was carried out using CARL ZESIS Model; EVO MA 15 scanning electron microscope. The surface of the samples was coated with conductive heavy metal such as gold.

### **Mechanical properties (ASTM D 882)**

Tensile properties of virgin PP, PP- Cu oleate and PP-Cu /S blended sample before and after UV exposure, with dimensions 150 x 25 mm were subjected to tensile tests as per ASTM D 882, using Universal Testing Machine (UTM), Lloyd Instrument Ltd, UK. A cross head speed of 500mm/min and gauge length of 50mm in both machine and transverse directions.

### **Optical properties**

Optical properties such as luminous transmittance and haze were studied for the PP- Cu oleate and Cu /S before and after UV exposure to find the effect of additive on the optical characteristics of the film. For measuring haze and luminous transmittance, The BYK Gardner Spectrophotometer was employed (ASTM D 1003).

## Elemental Analysis

The carbon content of the each test sample determined by elemental analysis by using Carlo Erbal model 1106 elemental analysis.

## ASTM D-5338: Test procedure

According to the procedure reported in the literature [20]. The percentage of biodegradation determined.

## RESULT AND DISCUSSION

### Fourier Transforms Infrared Spectroscopy (FTIR)

In FTIR spectra as shown in fig.1 (a) Cu 12- hydroxyl oleate and fig.1(b) starch to find the wave number and the peak assignments in table 2(a, b). The FTIR spectra of PP film containing Cu oleate and Cu /S as shown in figure 2(a, b). In fig.2 a, b (i) shows the virgin PP samples to be analyzed by IR were cut out from the films. In the IR spectra, special interest was focused on the following absorption peak: 973  $\text{cm}^{-1}$ , rocking vibration (-CH<sub>2</sub>-); 997  $\text{cm}^{-1}$ , rocking vibration (-CH<sub>2</sub>-); 1167  $\text{cm}^{-1}$  anti symmetric deformation (-CH<sub>3</sub>-); 1454  $\text{cm}^{-1}$  symmetric deformation (-CH<sub>2</sub>-); 1167  $\text{cm}^{-1}$ , Symmetric deformation (-CH<sub>3</sub>-); 1167 anti symmetric deformation (-CH-); 2929 symmetric stretching (-CH<sub>3</sub>-). In FTIR spectra PP with different percentage of Cu oleate and Cu / S additive is given in figure 2(a, b). A peak at around 1702-1750  $\text{cm}^{-1}$  corresponding to carbonyl group increase with increase of UV exposure to the films, increases the intensity which is due to the formation of new carbonyl groups on photo degradation involving the chain scission following the Norrish type 1 reaction.

In Fig.2 (c) The carbonyl index (CI) for the samples was calculated by normalizing the characteristics peak of PP at 1450  $\text{cm}^{-1}$  for all the spectra and calculating the area under the peak for this region and also 1710-1750  $\text{cm}^{-1}$ . It was found that at higher percentage before UV exposure slightly considerable peak carbonyl consist from the additive. But after 75 hour exposure to UV radiation, it's all formulation there was an obvious change in the corresponding peak which indicated the increase in the degradation rate with irradiation time. Fig.2 (d) shows the plot of CI as function of irradiation time PP- Cu oleate as well as Cu /S calculated and it's at higher percentage. In the case of carbonyl index from the plot PP- Cu oleate slightly higher than the Cu /S.

Table 1: UV cycle for photodegradation studies

Cycle	Irradiation $\text{W/m}^2$	Temperature ( $^{\circ}\text{C}$ )	Time (Hrs)
UV Irradiation	0.63	60	8
Condensation	-	50	4

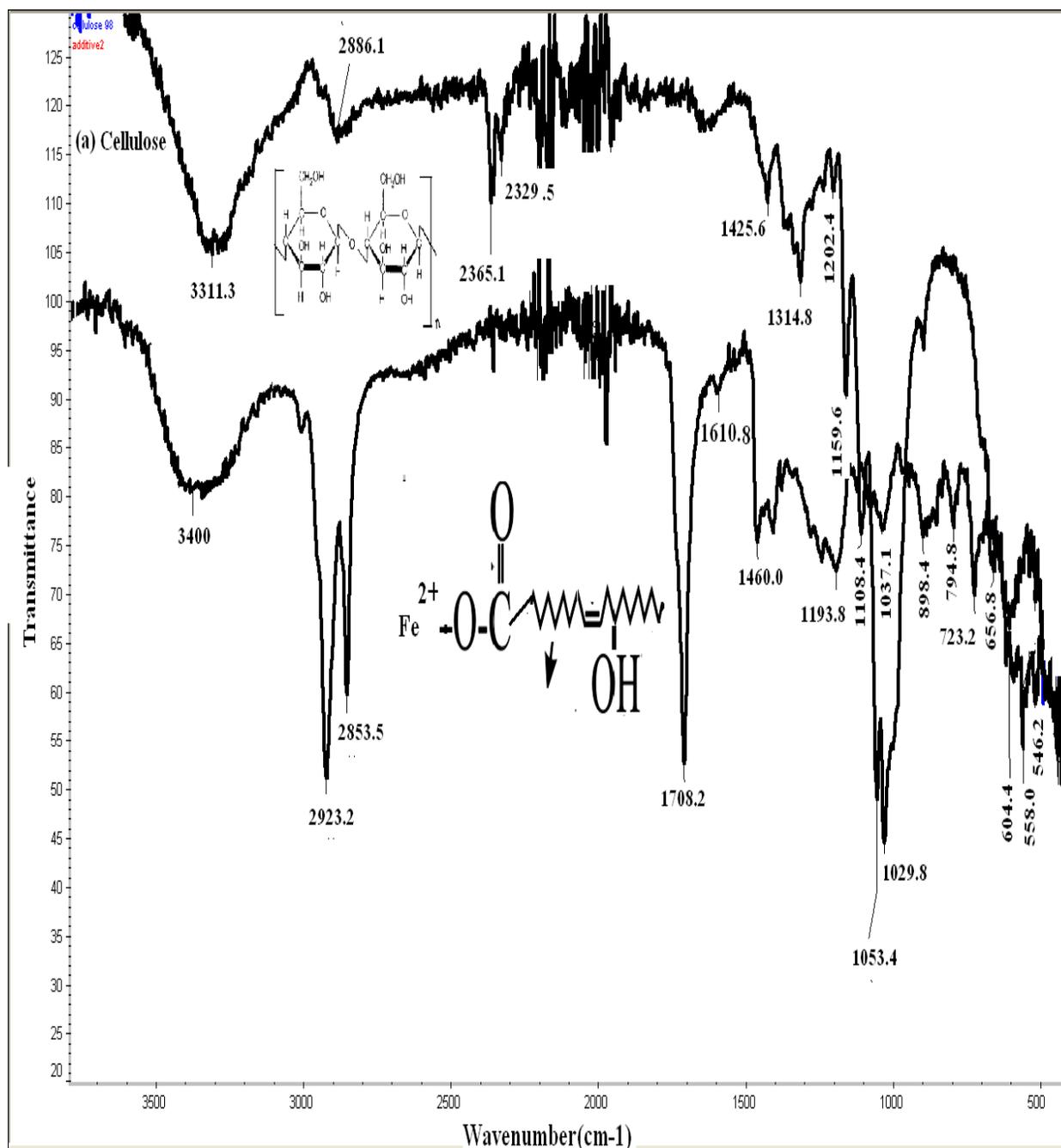


Fig.1 : Characterisation of FTIR spectra (a) Cu 12- hydroxyl oleate, (b) Starch

**Table 2: Characterization peak of FTIR spectra Cu oleate and Cu oleate/starch on UV exposure for 38 hours**

S.No.	Wave number (cm <sup>-1</sup> ) and their peak assignments (a) Cu-12 hydroxyl oleate	
1	1711	-C=O stretching
2	2723	-C-H stretching
3	2922	C-H stretching
4	1432	-C-C- stretching
5	1377	-CH <sub>3</sub> symmetric deformation
6	3312	O-H stretching (Carboxyl acid)
7	887.9 ,	=C-H alkenes
8	1034, 1192	C-O stretching
9	1610	-C=C- Stretching
10	720 & 667.2	C-H rock stretching & Copper complex stretching
S.No.	Wave number (cm <sup>-1</sup> ) and their peak assignments (b) Starch	
10	3240	O-H stretching (alcohol)
11	2365	C-H stretching
12	1034 ,1077	C-O stretching
13	1346	C-H rock (stretching)

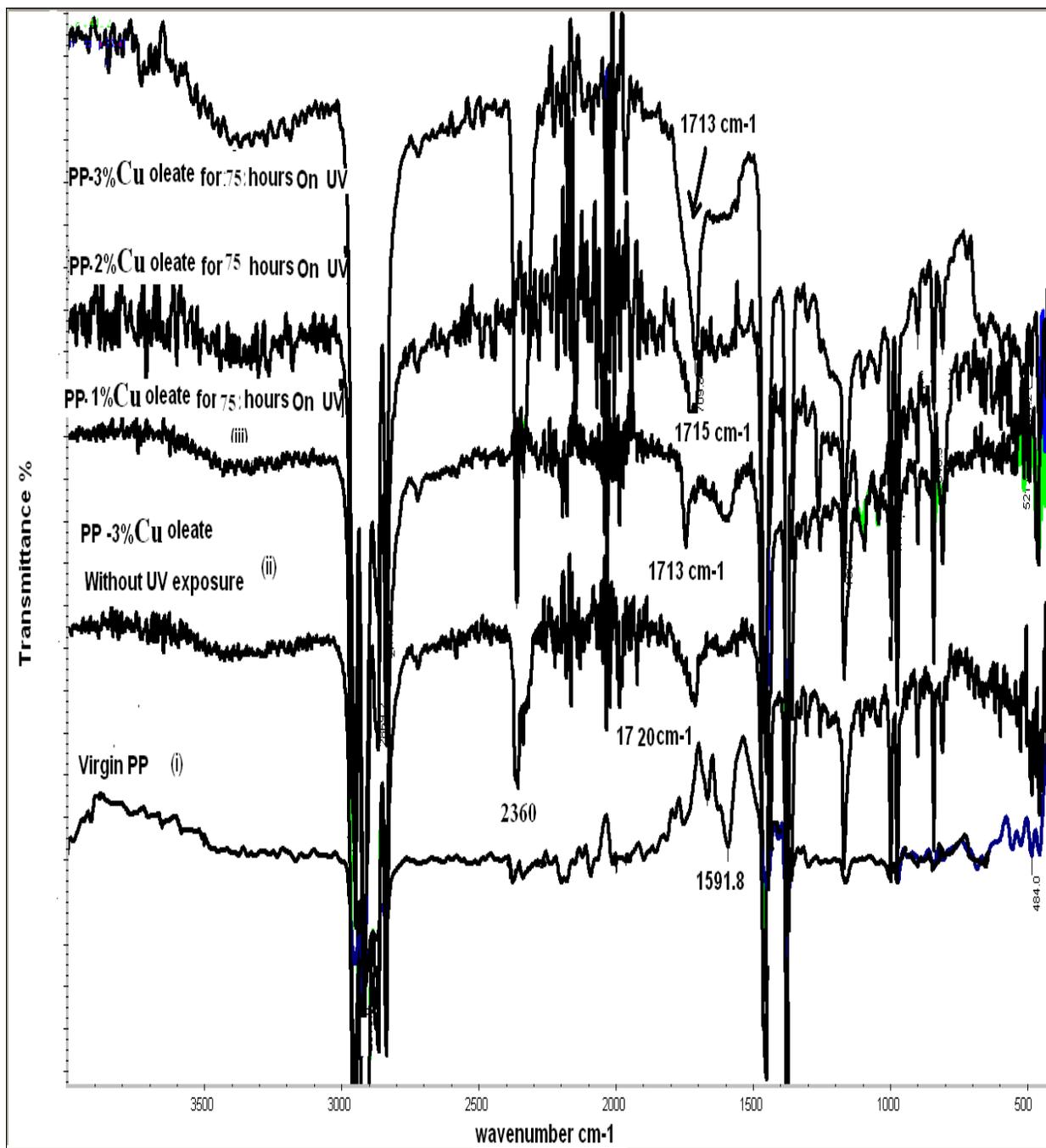


Fig.2(a)



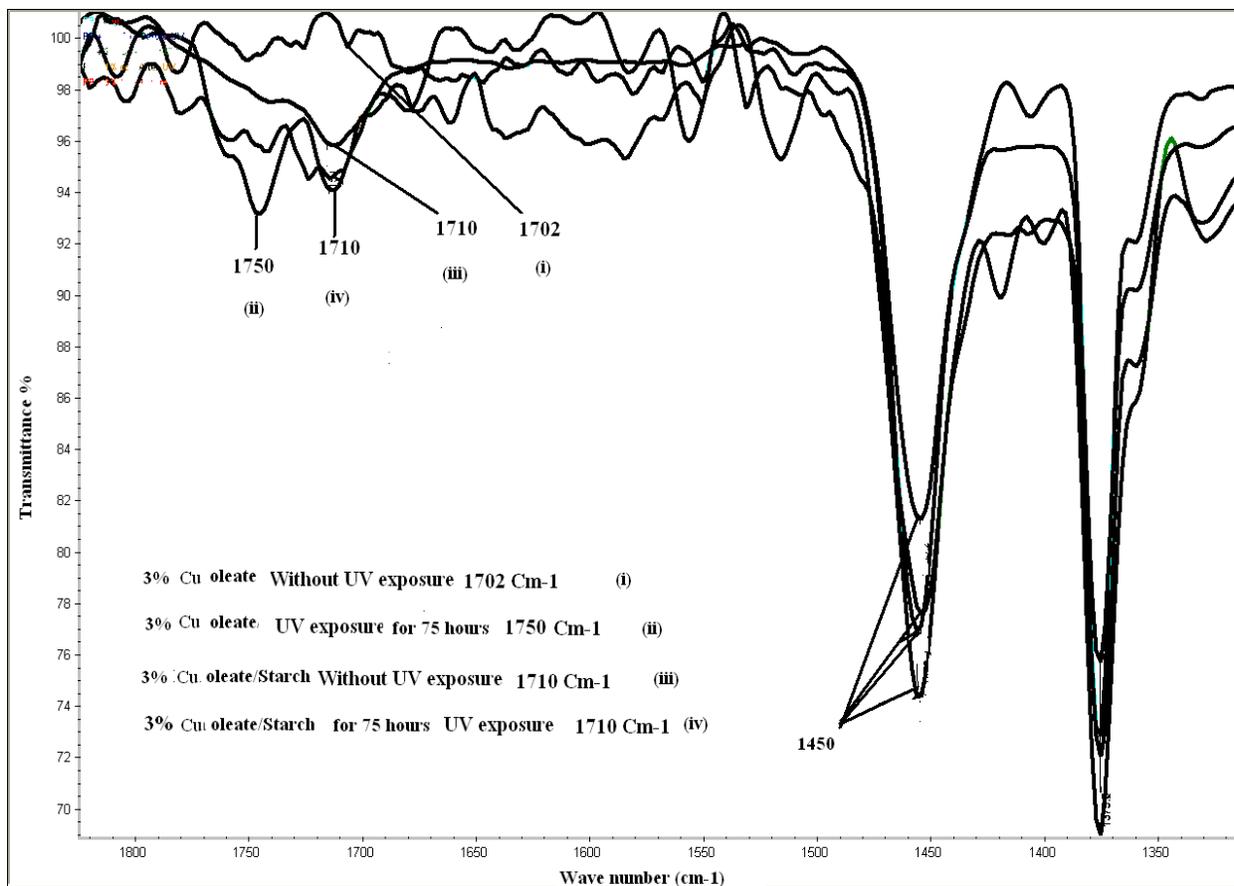


Fig. 2(c)

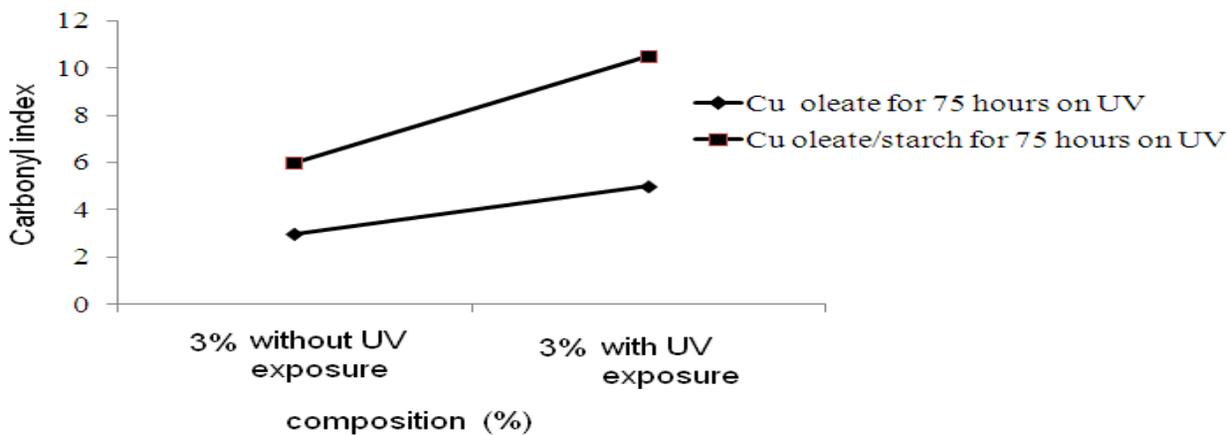


Fig. 2 (d)

**Fig.2 (a, b, c, d):** Comparison of FTIR spectra for different percentage (i)Virgin PP,(ii) without UV exposure 3% PP- Cu Oleate ,(iii) 1%, 2% and 3% PP- Cu Oleate with UV exposure (a), Comparison of FTIR spectra for different percentage (i) Virgin PP, (ii) 3% without UV exposure, (iii) 1%, 2% and 3% with UV exposure - Cu Oleate/starch (b), Comparison of carbonyl index 3% Cu oleate(i), Cu /S without UV exposure (ii), 3% Cu oleate (iii), 3% Cu /S (iv) with UV exposure (c), Change in carbonyl index due to photo oxidation on UV exposure at higher percentage for 75 hours (d).

Thermal properties

Differential Scanning Calorimetry (DSC)

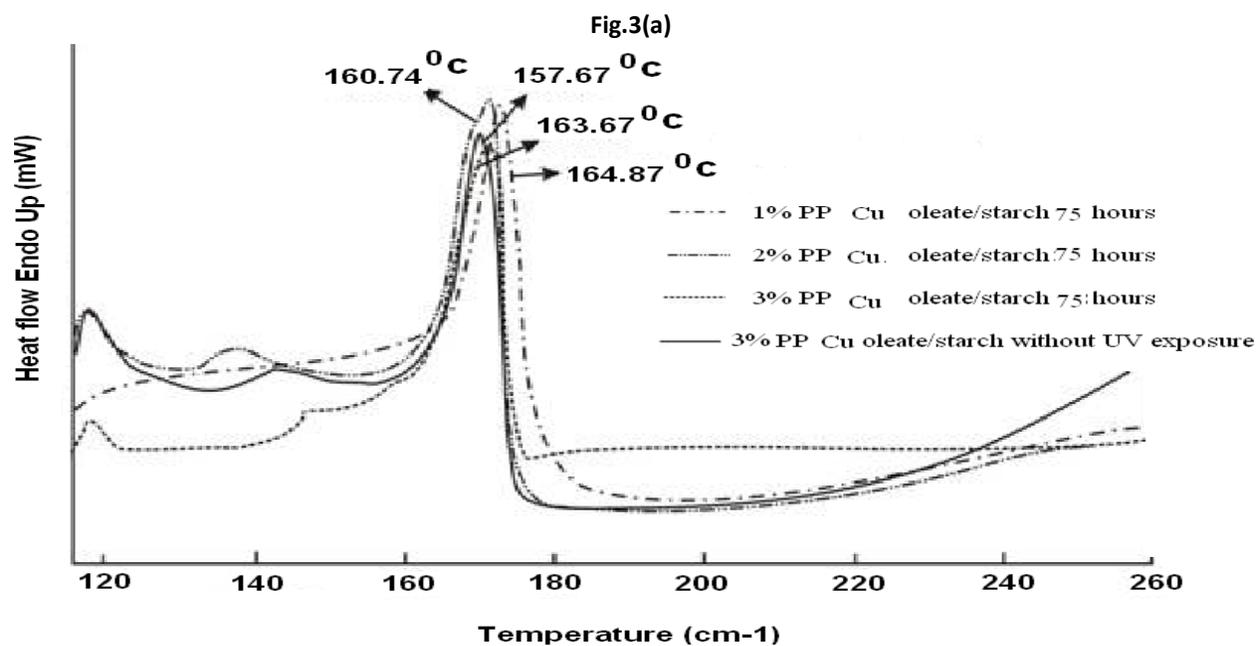
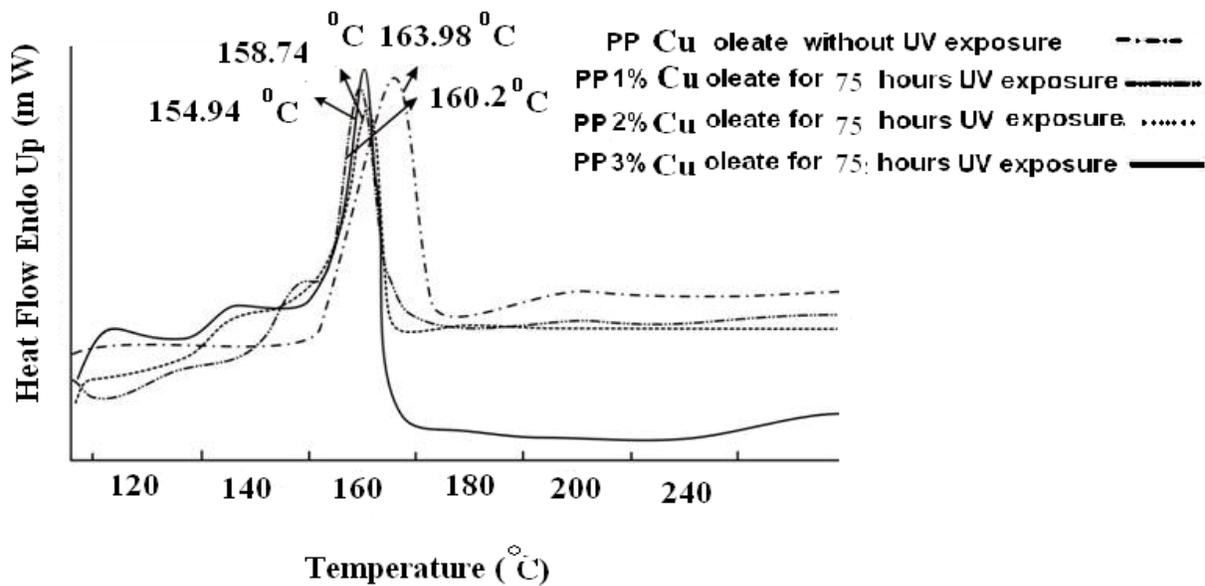


Fig. 3(b)

Fig.3 (a, b): Effect of melting point on UV exposure for 75 hours a) PP- Cu oleate, b) PP- Cu /S

The differential scanning calorimetry data pertaining to the melting point before and after exposure to accelerated UV is presented as shown in fig.3 (a, b). The virgin PP shows its melting point at 165.30°C. On the incorporation of Cu oleate, the melting point is found to change slightly due to the presence of additive in PP matrix. In case of the PP- Cu oleate samples exposed to UV for 75 hours a marginal decrease in the melting point from 163.98°C to 154.94°C as well as Cu /S from 163.98 °C to 157.67°C was observed. This implies that there is no much change in the melting point due to oxidation [8]. In this case of Cu oleate slightly lower than Cu /S due to Cu /S consist of more hydroxyl group interact within the polyolefins during on UV radiation but there is no change in the melting point without UV exposure at higher percentage due to when the UV exposure there is no strong interaction (Compatibilizing) within additive and polymer.

### Thermo gravimetric Analysis (TGA)

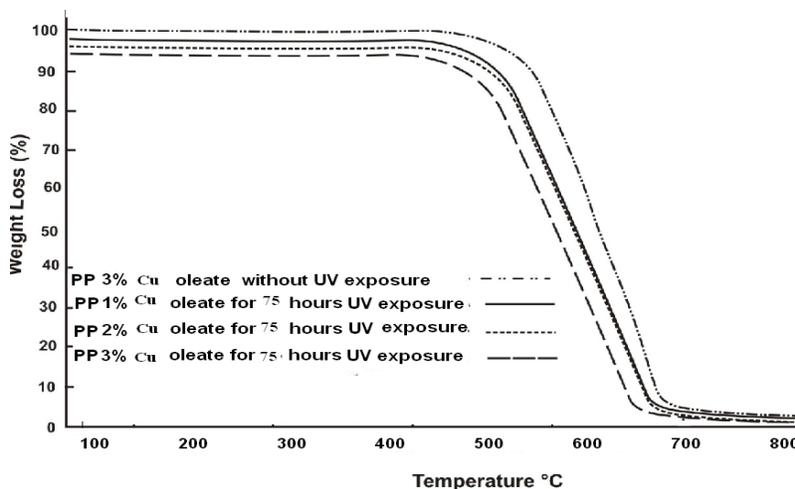


Fig. 4(a)

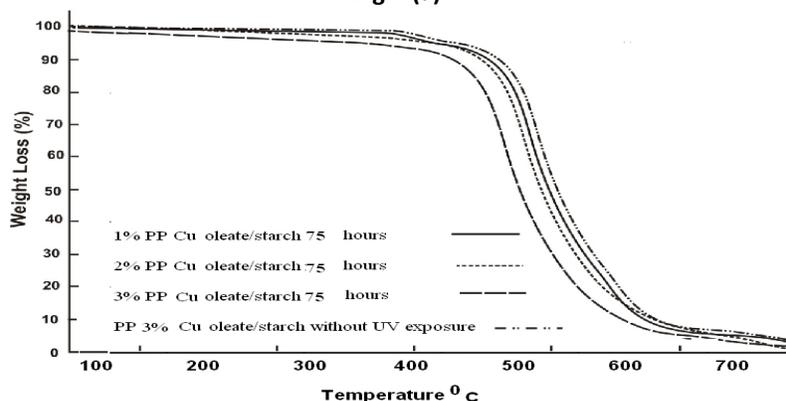


Fig. 4(b)

Fig.4 (a, b): Effect of thermal degradation on UV exposure for 75 hours a) PP- Cu oleate b) PP- Cu /S

The thermo gravimetric analysis of PP-Cu oleate and Cu / S additive is summarized as shown in figure 4(a, b). The results show that the initial decomposition temperature of PP after blending with Cu oleate from 427.1 to 403.0°C as well as from 427.1 to 415.0°C decreases

significantly for 75 hours in the presence of UV exposure. The increase in percentage of additive further decreases the initial decomposition temperature. In fact about 22°C and 12°C respectively, decrease in initial decomposition temperature was observed. Magagula et al, Marcelo A.G. Bardi et al [21, 22] said that a free radical may form on the polymer desired to the effects of heat. The free radical rapidly combines shield available oxygen to form a peroxy radical. This subsequently abstract labile hydrogen form a nearby polymer chain regenerating the special conglomerate release radical and resulting leverages a hydro peroxids. Under the influence of heat the latter cleaves to form two additional discharge radicals. This chain scission taking set down that leads to ruination of desirable polymer properties. Analyzing fig.4 (a, b) it is possible to observe that the PP- and Cu Cu oleate as well as Cu /S presented thermal behaviors, indicating that the exposure to UV radiation caused chain scission and cross-linking in PP. So thermal degradation affected by the increasing radiation dose in the presence of Cu oleate and Cu /S with the increasing percentage.

**Melt flow index (MFI)**

**Table 3: Effect of melt flow rate Cu oleate and Cu oleate/Starch on UV exposure for 75 hours**

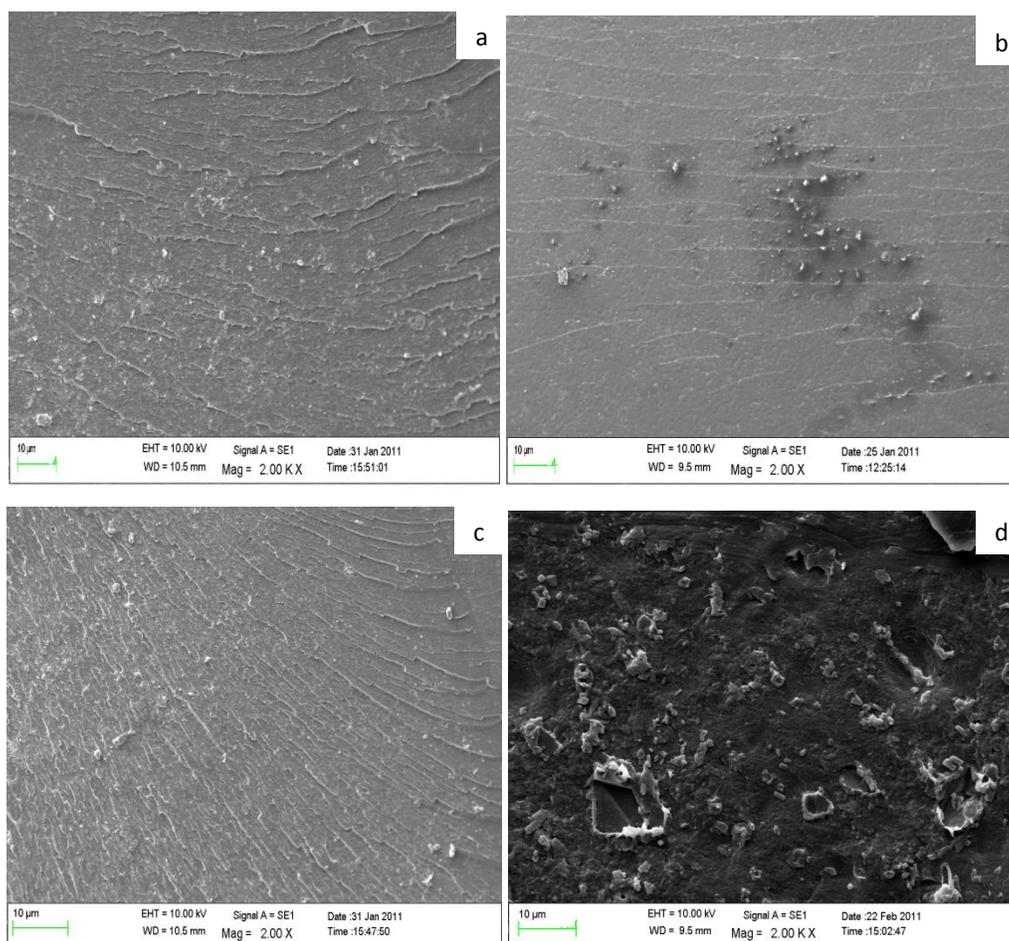
S. No	Sample Identification (a) PP- Cu oleate	Melt flow rate (MFI) g/10 min
1	PP Virgin	3.4
2	PP-3% Cu oleate without UV exposure	3.9
3	PP-1% Cu oleate with UV exposure for 75 hours	5.0
4	PP-3% Cu oleate with UV exposure for 75 hours	5.6
5	PP-5% Cu oleate with UV exposure for 75 hours	6.8
	<b>(b) PP- Cu oleate/Starch</b>	
6	PP-3% Cu oleate/starch without UV exposure	3.6
7	PP-1% Cu oleate/starch with UV exposure for 75 hours	4.7
8	PP-3% Cu oleate/starch with UV exposure for 75 hours	5.0
9	PP-5% Cu oleate/starch with UV exposure for 75 hours	6.3

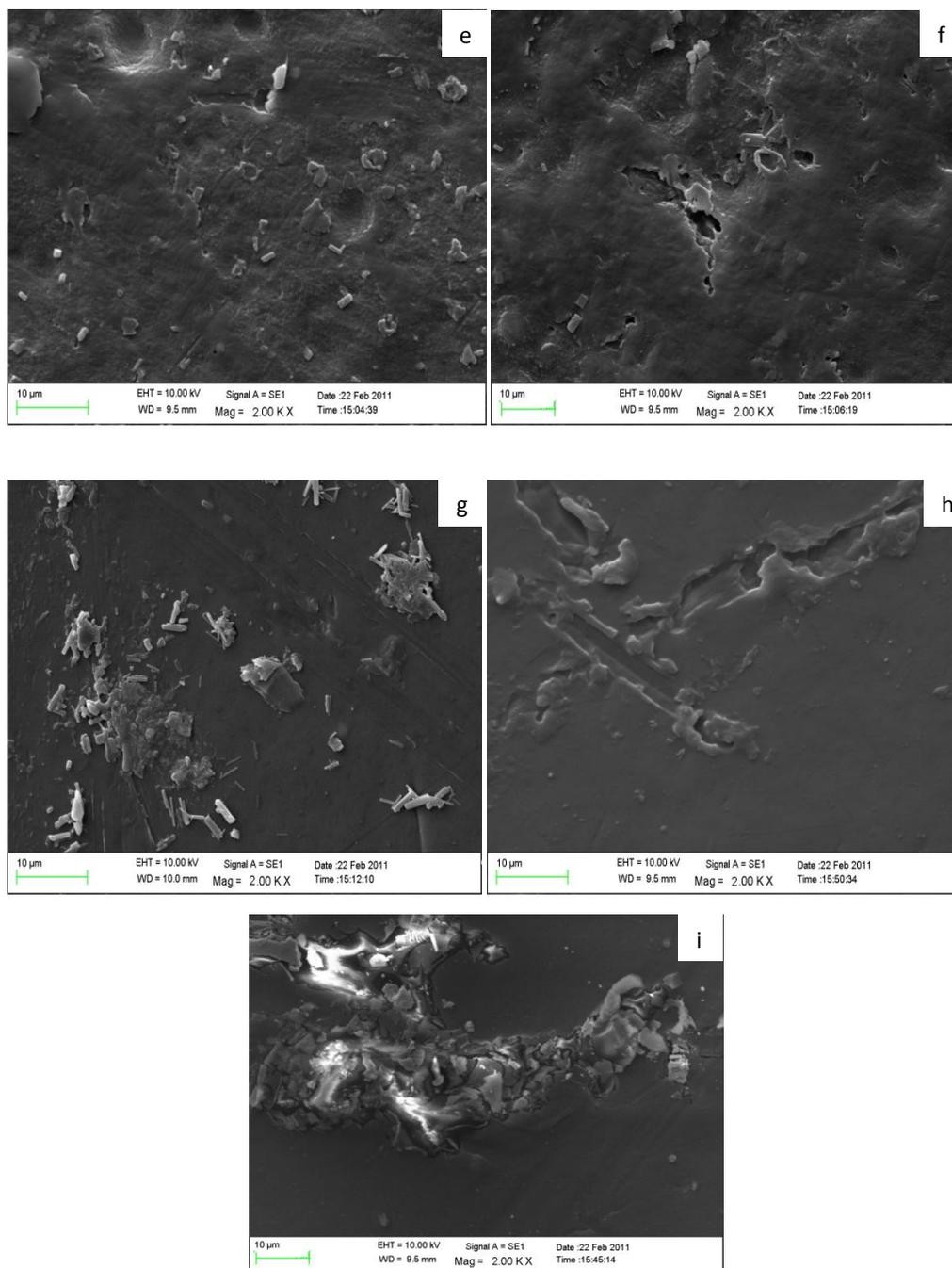
In table 3, the decreases in the molecular weight were confirmed by investigating the increase in MFI with exposure. Initial MFI of all the formulations was similar, indicating that the polymer does not undergo degradation due to processing. The MFI of PP 3.4 g/10min. containing Cu oleate increases slightly from 3.4 to 3.9 without UV exposure different percentage (1%, 3% and 5%) while that of film containing at higher percentage additive from 3.0 to 6.8 g/10 min. after UV exposure for 75 hours. Similar way Cu /S increases slightly from 3.6 to 6.3 on UV exposure for 75 hours.

**Scanning Electron Micrograph (SEM)**

The analyses of SEM microphotographs of the fracture surfaces samples of PP- Cu oleate and Cu /S, before and after exposure to UV radiation, illustrated in Fig. 5(b-i). Scanning Electron Micrographs of the samples at magnification of 2000x. Fig. 5(a) as is apparent from the surface of nondegraded PP is smooth, without fractured and free from imperfection. The

SEM micrographs of PP-3% Cu / S blended film show the uniform dispersion of additive in the polymer matrix as shown in fig.5 (b, c). The scanning electron micrographs of fractured surface of films after UV exposure given in fig.5 (d-i) show the brittle mode of fracture. Fig.5 (d-i) shows the SEM degradation at magnification of 2000 x and its extent of damage was much more distinct in the sample containing in Cu /S as compared to neat PP. The films had a smooth surface before irradiation it can be seen that the surface agglomerates were formed which could be due to the photodegradation involving chain scission and deterioration of molecular chains. More surface agglomerations could be seen in the case of PP- Cu oleate and PP- Cu / S indicating the faster rates of photodegradation. Also, the brittleness of the surface increases with increasing the exposure time and percentage of additive concentration.





**Fig.5(a, b):** Virgin PP on Morphology (a), Effect of oleate on Morphology 3% PP-Cu oleate (b), 3% PP- Cu /S (c), Effect of oleate on Morphology 1% PP- Cu oleate (d) 1% PP- Cu /S (e), Effect of oleate on Morphology 2% PP- Cu oleate (f) 2% PP- Cu /S(g), Effect of oleate on Morphology 3% PP- Cu (h) PP- Cu /S(ii) (i) on UV exposure for 75 hours.

### Mechanical properties evaluation

The tensile strength and elongation at break data of PP film with Cu oleate and Cu / S before and after UV exposure are presented in the figure 6(a, b). The tensile strength of PP

virgin sample decreases with the incorporation of Cu oleate and Cu /starch in the concentration of 1%, 2% and 3%. The virgin PP shows on tensile strength reduced in the machine direction from 50.17 to 39.45 and 40.45 to 34.65 respectively, transverse directions from 48.25 to 36.65 and 47.35 to 34.65 respectively, on UV exposure while elongation at break reduced in the machine direction from 20.5 to 13.7 and 17.02 to 13 from respectively, reduced in the transverse direction from 131 to 68 and 120.1 to 59 respectively. There was a considerable decrease in the tensile strength and elongation at break on exposure of the films with additive to UV radiation. The Cu oleate and Cu / S based additives show considerable decrease in the tensile strength and elongation at break. The deterioration in mechanical properties of films was observed in both the machine direction and transverse direction, but the rate of deterioration was high in transverse direction. Also, increasing the concentration of additive from 1% to 3% increases the degradation. PP films with Cu oleate and Cu / S additive show decrease in tensile strength and elongation and ultimately become brittle when exposed to UV radiation for 75 hours and its all the formulation become brittle on UV exposure. This could be due to the fact that in the photolysis process of Cu oleate and Cu / S, the decrease in tensile and elongation is basically because of the polymer chains in the presence of sensitizer take up oxygen and this leads to the formation of hydro peroxide which breaks down to give low molecular weight oxygenated products [23, 8]. In this case Cu oleate, polymer chain scission slightly higher than Cu /S due to more hydroxyl group interacts within the polymer to improve the strength.

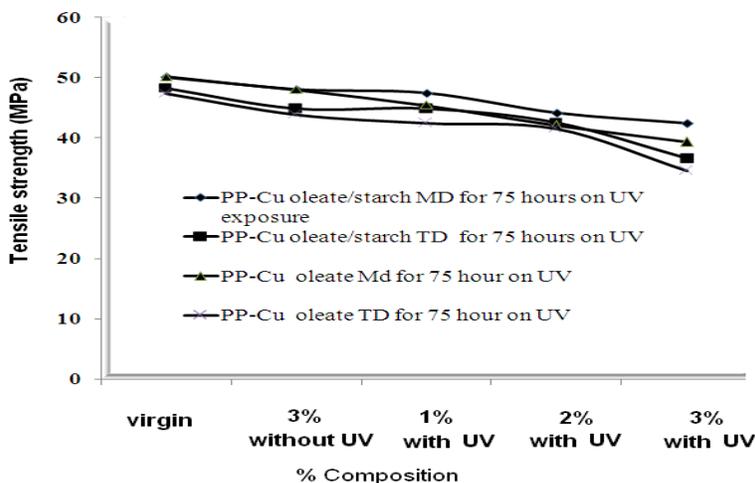


Fig.6 (a)

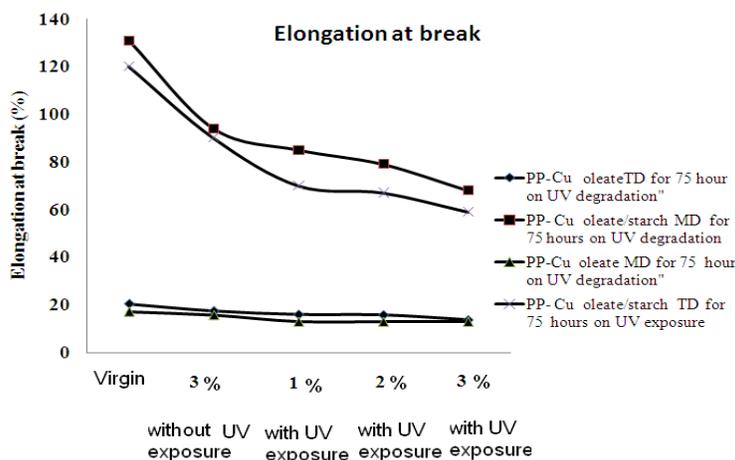


Fig.6(b)

Fig.6 (a, b): The effect of UV exposure on tensile strength (Machine and Transverse direction) for 75 hour in polypropylene film (a), The effect of UV exposure on elongation at break (Machine and Transverse direction) for 75 hours in polypropylene film (b)

**Optical property**

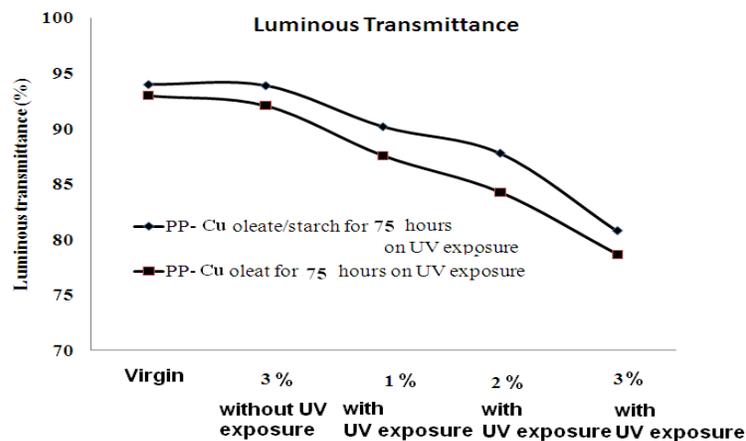


Fig.7 (a)

The results of optical properties of Cu oleate and Cu / S blended PP before and after exposure to UV radiation are given in fig.7 (a, b). It is evident that with the increase in additive concentration there was a decrease in transmittance level in the range from 94 to 80 and 93 to 78 respectively, and also increase in the additive concentration there was an increase in haze in the range from 19 to 48 and 17 for 45 hours and its all the formulation become brittle film when exposed to UV exposure. This is due to the carbonyl formation in the process of photo oxidative degradation of PP film.

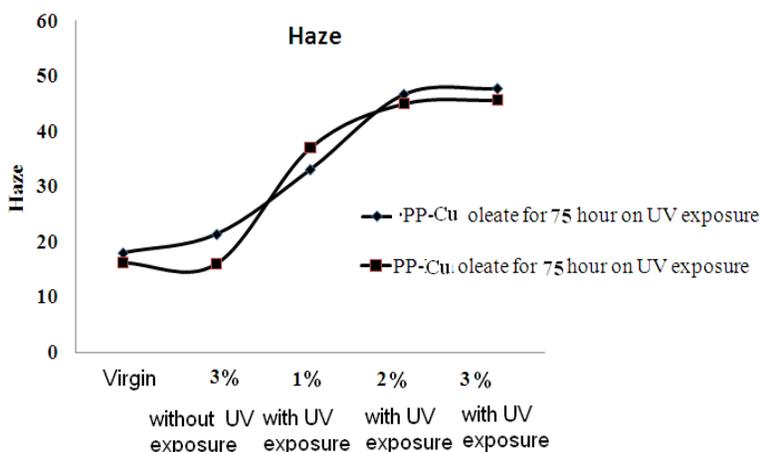


Fig. 7(b)

Fig.7 (a, b): The percentage of biodegradation of a) PP- Cu oleate at higher percentage 19% b) PP- 3% Cu oleate/starch-24%, Cu oleate and Cu /S for 45 days and it's both reference cellulose-80% in ASTM D 5338

In case of the films containing three additives possess low transmittance and high haze. As the UV exposure time increases the % haze and luminous transmittance decreases which could be due to the faster rate of degradation.

### Elemental Analysis

Table 4: Elemental analysis percentage of Carbon, Hydrogen and Nitrogen

Sample	Cellulose (%)	1% Cu oleate PP (%)	3% Cu/S PP (%)	5% Cu/S PP (%)	1% Cu /S PP (%)	3% Cu/S PP (%)	5% Cu/S PP (%)	Compost (%)
Carbon	84.47	86.22	84.36	84.36	86.20	84.00	84.35	14.32
Hydrogen	14.96	12.88	14.89	14.43	12.90	15.25	14.44	1.74
Nitrogen	0.57	0.90	0.75	1.21	0.90	0.75	1.21	1.54

C, H, N elemental analysis reported in table 4. Cu oleate and Cu /S exhibit of Cu composition 9.67 %.

### Biodegradation Testing

In fig.8 (a, b) shows conditions of reaction mixtures: Organ of compost; livestock excrement, municipal and Vegetable waste used the method used for the determination of the biodegradability of the polyolefin's was based on the International Standard (ASTM D 5338-98) that measures the evolved carbon dioxide (CO<sub>2</sub>) amount from both the blank vessel without a sample and the sample vessel including a 10 g PP- Cu oleate and Cu / S samples. According to the reference [20] ASTM D 5338 test procedure. Fragments occur progressively in the biodegradation of the photodegraded films. Moreover, the biodegradation test results reveals that the Cu oleate and Cu / S shows 19% and 24% respectively, of biodegradation on

photodegraded PP films when observed at the end of 45 days. In this case reaction condition in composting vessel, reaction temperature:  $58 \pm 2^\circ\text{C}$ , Dry solid (%): 52%, Volatile solid (%): 20%, Air flow rate; 100 ml/min, Test duration (day): 45 day, Reference material: Cellulose, Volume of reaction vessel: 2000ml, Moisture percentage in compost: 28 %, Composting vessel (material with test medium) before and after biodegradation at  $\text{pH}$  range 7.6 - 7.8.

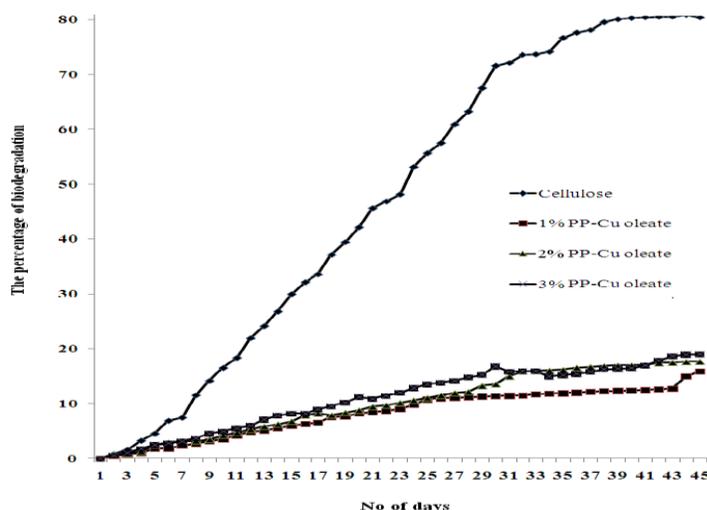


Fig. 8 (a)

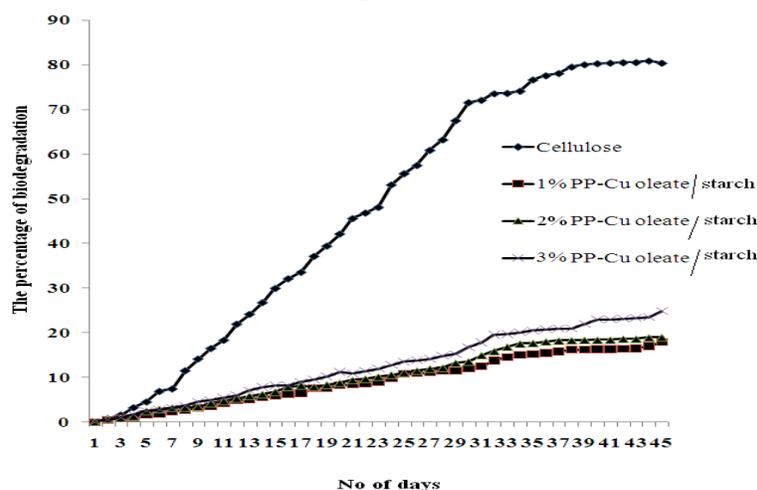


Fig.8 (b)

### CONCLUSION

The rate of photodegradation on PP film is very high at higher concentration of Cu oleate and Cu / S. PP films containing 3% Cu oleate and Cu / S has shown highest degradation in mechanical, thermal properties on UV exposure. In the FTIR analysis shows that a peak at around  $1702\text{-}1750\text{ cm}^{-1}$  corresponding to carbonyl group and carbonyl index of both additives were observed for PP films with different formulation of additive. The SEM micrographs indicate the fractured surface and brittle mode on films after the effect UV exposure. Fragments occur progressively in the biodegradation of the photodegraded films. Moreover,

the biodegradation test results reveals that the Cu oleate and Cu /S shows 19% and 24% respectively, of biodegradation on photodegraded PP films when observed at the end of 45 days. It can be concluded the above characteristics photodegraded PP film, the polypropylene chain scission Cu oleate slightly higher than the Cu /S on UV exposure for 75 hours but the percentage of biodegradation on photodegraded film the Cu /S slightly higher than the Cu oleate.

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