

# Preparation and Structural Characterization of Melamine–Methylurea–Formaldehyde Resin and Its Blends Separately with Ethyl Cellulose, Starch, Teakwood, and Almond Shell Powders by $^{13}\text{C}$ NMR, IR, TGA, and SEM Techniques

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**ABSTRACT:** Melamine–methylurea–formaldehyde (MMUF) polymer resin was prepared by condensation polymerization and the molecular weight was determined by viscometry. It was blended with dried starch and ethyl cellulose, and cellulose and hemicellulose-rich teakwood and lignocellulose-rich almond shell dried powders separately. The structural characterization of resin with  $^{13}\text{C}$  NMR and IR spectroscopy, and morphological studies on the resin and blends were undertaken with thermogravimetric analysis (TGA) and Scanning Electron Microscopy (SEM). The micrographs of the starch and ethyl cellulose have found resemblance with pure resin while

teakwood and almond powders have been found to differ in structure from starch and ethyl cellulose due to crack masks in the bulk structure. The different weight loss at different temperatures of the blends was observed, which proves that blending materials do control the weight loss of resin with TGA or curing. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 92: 3437–3446, 2004

**Key words:** oligomer morphology; oxidative degradation; thermogram

## INTRODUCTION

For a long time the preparation and characterization of polycondensation resins like UF (urea-formaldehyde) and MF (melamine-formaldehyde) have been of interest. The preparation and properties of them can be correlated to the synthesis of urea by Whohler,<sup>1</sup> in 1824, and formaldehyde by Burtlerov, in 1859. Later, Tollens,<sup>2</sup> in 1884, first attempted to investigate the structure of resin products of urea and formaldehyde reactions, and since then, several workers<sup>3–11</sup> have reported enormous studies on them. Similarly, the studies and various industrial applications of MF were investigated.<sup>12,13</sup> Later, the condensation of reaction of urea and melamine with formaldehyde were carried out, and various hydroxy compounds named oligomers were detected.<sup>14</sup> It is of interest to know that an increase in the formation of oligomers is observed when the pH of the initial solution decrease.<sup>15–18</sup> Such chemical behavior of these reagents has been considered an important feature of aminoplastic kinetics.<sup>19</sup> Cost-wise urea is found cheaper than melamine, and reaction-wise simple, so efforts were made to synthesize melamine–urea–formaldehyde (MUF) resins on

copolymerization of urea, melamine, and formaldehyde, which have been obtained with wide application. Several kinds of physicochemical properties of the polycondensates were tested on molding compounds of MUF.<sup>20</sup> In comparison to UF and MF resins, the processing of MUF has been found superior.<sup>21,22</sup> In search of such properties of resins an attempt was made to prepare melamine–methylurea–formaldehyde (MMUF) resins, which have been blended with naturally occurring cellulose materials from woods. Basically, MMUF has been an extension of MUF. In general, the composition of wood is 50% cellulose, 15–30% hemicellulose, 15–35% lignin, and 5–30% W by W ash and extractives. So teakwood and almond shell powders have been chosen for blending to obtain new composite materials with qualities compatible with an industrial applications. The almond shell also belongs to a category of wood. The structures of resins have been investigated with IR and  $^{13}\text{C}$  NMR, but blends in addition to IR were subjected to thermal gravimetric analysis (TGA) and scan electron microscopy (SEM) for bulk structure elucidation. In general, the large quantities of UF and MF resins are used as adhesives for paper binding and in the production of molding compounds<sup>23,24</sup> and foams<sup>25</sup> in pure form. These resins have been of current interests, as hydroxymethyl compound form oligomers, which further form resin networks. Phenol–urea–formaldehyde<sup>26</sup> phenol–urea, and melamine–formaldehyde, urea furfuryl alcohol–formaldehyde<sup>27,28</sup> copolycon-

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densate resins are prepared in industries for various applications. But due to the superior quality of resin and simplicity in preparation, the melamine–urea–formaldehyde (MUF) polycondensates are in use as adhesives for pulpwoods,<sup>29–35</sup> IR<sup>36</sup> and NMR<sup>37–42</sup> spectroscopy have investigated their structures. An enormous studies on MUF,<sup>43–47</sup> melamine–formaldehyde,<sup>48</sup> melamine–urea, and urea–formaldehyde<sup>49</sup> have been conducted in detail, while studies on MMUF resin preparation and its blends with cellulosic items have not been reported. This manuscript reports the structural, morphological, and thermal stability investigations on the resin and its blends. This is aimed to characterize the adhesive resins with high strength and extensive applications. The purpose of blending the resin is targeted to develop ecofriendly composite materials and coatings for metallic surfaces. This is focused to introduce degrading properties in thermoset polymer by naturally occurring cellulosic materials. This approach of such studies is conducted first time to explore greater applications that could also encourage the preparation of resins with di- and trimethyl urea, which may be of some use in the field of resin.

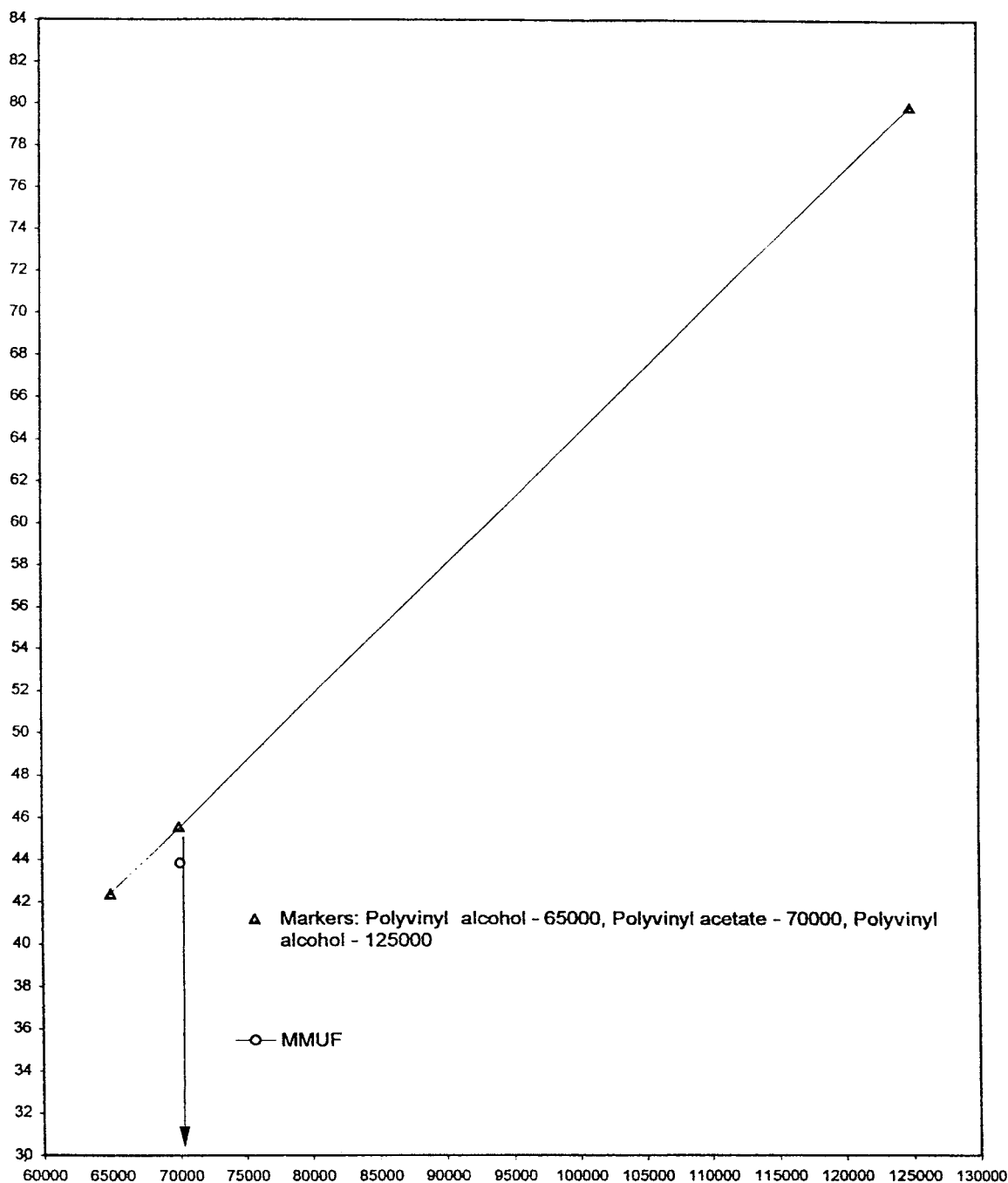
## EXPERIMENTAL PROCEDURES

Melamine and methylurea (AR, BDH), and 40% formalin solution (E. Merck) were used. The melamine (m.p. 300°C) at 150 and methylurea (m.p. 95°C) at 80°C were kept overnight in an oven, and thereafter vacuum desiccator filled with P<sub>2</sub>O<sub>5</sub> for 24 h for complete dryness. The powders of 100 μ of teakwood and almond shell, and starch and ethylcellulose (AR, BDH) were kept in an oven overnight and thereafter in a P<sub>2</sub>O<sub>5</sub>-filled desiccator for drying. The initial melamine : methylurea : formalin molar ratio was 1 : 5 : 1 and taken in a 500-mL capacity round bottom Borosil flask socketted with three necks. In the central socket a reflux condenser was fitted and a thermometer with a pocket in the second. The third neck is 10' long and in its socket a 5-mL syringe is fitted, and a 10'-long stainless steel needle of 2 mm i.d. and 13' in length was fitted with a syringe to suck out the sample from the reaction mixture. The flask was heated in oil (groundnut) bath @ 2°C min<sup>-1</sup> for polymerization, and after every 5°C rise in temperature an aliquot of reaction mixture was sucked out to a watch glass for a checking of adhesives of the undergoing product. Likewise, the sampling is done up to about 55 to 60°C where reaction mixture starts developing a greater adhesive strength. This further intensifies after 75°C, and at about 80°C the heating is stayed for 10 to 15 min, and after this, the sample is quenched by placing the flask with contents to an ice-cold water bath. After 35 to 45 min the product becomes a rigid and whitish resinous material; it seems that melamine and methy-

lurea methylols named oligomers are formed on heating up to 80 ± 1°C. The quenching seems to stop crosslinking of melamine and methylurea methylols. The reaction was comparatively slow at room temperature and the structure of the formed product depends on the composition of the solution and the duration of the reaction.

The molecular weight of the prepared MMUF resin was determined viscometrically using a low shear Ubbelohde viscometer. As the resin was found soluble in water, the viscosity measurements were carried out on very dilute aqueous solutions of it at 25 ± 0.01°C, the intrinsic viscosity,  $[\eta]$  derived from viscosity values were fitted to the Mark-Houwink equation<sup>45</sup> to calculate the weight-average molecular weight ( $M_w$ ). The  $[\eta]$  was obtained as intercept ( $[\eta] = \eta_{sp}/c, c \rightarrow 0$ ) of  $\eta_{sp}/c$  vs.  $c$  plot. Here,  $\eta_{sp}$  and  $c$  stand for specific viscosity and concentration of resin solutions. Firstly, the  $\eta_{sp}/c$  vs.  $c$  plots of the aqueous and dilute solutions of polyvinyl alcohol, polyvinyl acetate, and polyvinyl alcohol as markers with molecular weights of 65,000, 70,000, and 125,000 are accessed separately. The markers are chosen as they belong to the category of condensation resin, and their  $[\eta]$  values were fitted against their molecular weights to draw a calibration curve (Fig. 1), where the value of  $[\eta]$  of resin was put on the curve and 70,100 g mol<sup>-1</sup> weight was estimated.

The fine powder of the blending materials was made of about 100 μ by an electric grinder and completely dried. The resin : blending material ratio was 4 : 1 in grams, and each material was blended separately with resin. The weighted amounts of blending materials were taken in 20-mL capacity beakers and resin was added to each and mixed to an homogenous state with a trident and flat-type tipped glass rod stirrer fitted to an electric wiper motor. The purpose of blending with the structure of starch and ethyl cellulose is to load the resin framework with purely degradable structure. The major content of teakwood powder was cellulose and hemicellulose with a little percent of lignin, while of almond shell was lignocellulose and pectin. The proton-decoupled <sup>13</sup>C NMR on resin was recorded with a Bruker 300 MHz Spectrometer in DMSO-d<sub>6</sub> using nondeuterated DMSO as an internal standard. The thermogravimetric analysis for the resin and the blends was done with a Thermal 2100 module 951 Thermogravimetric analysis, TA Instruments (New Castle, DE) in an atmosphere of nitrogen. The samples of approximately 10-mg weight were heated at a rate of 10°C min<sup>-1</sup> in a crucible positioned on a quartz beam attached to an automatic recording balance in a furnace connected to a taut-band suspension electromechanical transducer. The horizontal quartz beam was maintained at a null position by current flowing through the transducer coil of an electromagnetic balance. The movements of the beam were maintained by photosensitive diodes,



**Figure 1** Calibration curve for MMUF-average molecular weight determination with Ubbelohde viscometer at 25°C. Y-axis: intrinsic viscosity in liter mol<sup>-1</sup> and X-axis: molecular weight of markers in g mol<sup>-1</sup>.

which acts as a position sensor, and a change in weight of the sample causes a deflection of the beam. It was detected by one of the photodiodes and the beam was then restored to the original null position by a feedback current sent from the photodiodes to the coil of the balance. The current is proportional to the change in weight of the sample and  $\Delta w/\Delta t$  was computed by software attached to a TG instrument. With 0.01 cm<sup>-1</sup> resolution, FTIR spectra for the resin and the blends were recorded in the form of a thin film

within a KBr disc using a Nicolet Protégé-460 Spectrometer, and their morphological features were observed using a Stereoscan 360 Cambridge scattering electron microscope.

## RESULTS AND DISCUSSION

IR and <sup>13</sup>C NMR spectra have investigated the structure of MMUF resin. Several bands have been found,

TABLE I  
Structure of Oligomers of Resin with Chemical Shift ( $\delta$ ) in ppm, the H of  $-\text{NH}_2$  (Amino Groups) of Melamine and Methylurea Is Substituted by  $\text{CH}_2(\text{OH})_2$  (Methylol), This Increases the Numerical Value of  $\delta$  ppm

Figs.	Bands	Triazine (melamine) nucleus and its substitutions			
		Name	structure	$\delta$	
2(b)	1	Triazine nucleus	$\text{me}=\text{C}-\text{NH}_2$	167.4	
	2	Monosubstitution	$\text{me}=\text{C}-\text{NHCH}_2-$	166.3	
	3	Disubstitution	$\text{me}=\text{C}-\text{N}(\text{CH}_2)_2$	166.0	
	Methylurea residues				
	4	Monosubstituted	$\text{H}_2\text{NCON}(\text{CH}_3)\text{CH}_2\text{O}-$	159.9	
	5	Disubstituted	$>\text{N}-\text{H}_2\text{C}-\text{HNCON}(\text{CH}_3)\text{CH}_2\text{N}<$	159.2	
	6	-d-	$-\text{O}-\text{CH}_2\text{NHCON}(\text{CH}_3)\text{CH}_2\text{N}<$	159.0	
	7	-do-	$-\text{O}-\text{CH}_2-\text{NHCON}(\text{CH}_3)\text{CH}_2\text{O}-$	158.6	
8	Trisubstituted	$-\text{OH}_2\text{C}$ $\begin{array}{l} \diagdown \\ \text{N}-\text{CON}(\text{CH}_3)\text{CH}_2\text{O}- \\ \diagup \end{array}$	158.1		
2(c)	1	C with N	$=\text{C}-\text{N}$	26.4	
	2	$\text{CH}_3$ of methylurea	$-\text{CH}_3$	33.6	
	3	$\text{DMSO}_d_6$	$(\text{CH}_3)_2\text{SO}$	40.0	
	4	Methylene units in linear chain	$-\text{OCHN}-\text{CH}_2-\text{NHCO}-$	45.7	
	5		$-\text{OCHN}-\text{CH}_2-\text{NH}-\text{C}$	46.0	
	6		$\text{C}-\text{HN}-\text{CH}_2-\text{NH}-\text{C}$	47.0	
	7	Methylene units in branch	$-\text{OCN}(\text{CH}_3)-\text{CH}_2-\text{NH}-\text{CO}-$ or $\text{C}-\text{N}-\text{CH}_2\text{NH}-\text{C}$	54.1	
Methylol groups on					
8	Secondary N atom	$-\text{OH}_2\text{CHNCON}(\text{CH}_3)-\text{CH}_2\text{OH}$	63.9		
9	-do-	$\text{C}-\text{NH}-\text{CH}_2\text{OH}$	64.2		
Methylate groups on					
10	Secondary N atom	$-\text{CON}(\text{CH}_3)\text{CH}_2\text{O}-\text{CH}_2-\text{N}(\text{CH}_3)\text{CO}-$ or $\text{C}-\text{NHCH}_2\text{O}-\text{CH}_2-\text{NH}-\text{C}$	67.6		
Methylol groups on					
11	Tertiary N atom	$\text{HOH}_2\text{C}$ $\begin{array}{l} \diagdown \\ \text{N}-\text{CON}(\text{CH}_3)\text{CH}_2\text{O}- \\ \diagup \end{array}$ or $\text{HOH}_2\text{C}$ $\begin{array}{l} \diagdown \\ \text{N}-\text{C} \\ \diagup \end{array}$ $-\text{OH}_2\text{C}$	71.4		

$^{13}\text{C}$  NMR in  $\text{DMSO}_d_6$  solvent. me = melamine.

and NMR shifts are given in Table I. These seem to be assigned to the substitutions of amino ( $-\text{NH}_2$ ) groups of melamine and methylurea forming melamine ( $=\text{C}-\text{NH}-\text{CH}_2\text{OH}$ ) and methylurea ( $>\text{NCONH}-\text{CH}_2\text{OH}$ ) methylols, respectively, and the monosubstituted amide group ( $-\text{CONH}-$ ) of methylurea have been detected. The previous<sup>46,47</sup> analytical result on MUF (melamine urea formaldehyde) has also shown, that these resins contain methylol, methylene, dimethylene ether, and amide groups. Such kinds of structural complexities are due to three functional groups

of MMUF, two of formaldehyde, and six of melamine, and make feasible a large number of likely structures. The viscometric method was used to determine molecular weight, and this gives average values only. A separation of the products into its individual component, for example, by paper and TLC, is subject to the damage of the structures being changed during analysis. Hence, IR and  $^{13}\text{C}$  NMR have been found suitable for structure elucidation. The same method has been used earlier to study formaldehyde solution. The chemical shifts ( $\delta$ ) of the  $^{13}\text{C}$  NMR signals are much

more sensitive to resolve a minute structural difference than of  $^1\text{H}$  NMR, as  $^{13}\text{C}$  detects a signal of the carbon atom that carries an identifiable proton with more certainty.

The proton decoupled  $^{13}\text{C}$  NMR spectra recorded on the resin in  $\text{DMSO-d}_6$  are depicted in Figure 2(a) to (c), and the signals were compared with the spectra of melamine formaldehyde and urea formaldehyde resins, and the assigned values are given in Table I. It is seen that signals are displaced into the weak field upon substitution of  $-\text{NH}_2$  groups of melamine and methylurea by  $\text{CH}_2(\text{OH})_2$  (methylene hydroxide) as a binder.

The complete  $^{13}\text{C}$  NMR is reported in Figure 2(a), and its expanded form with sensitive functional zones from 25 to 75 and from 156 to 175 ppm in Figure 2(b) and (c), respectively. This clearly depicts that the melamine and its substituted hydroxy products fall under a low field and of methylurea, a high field. A low field gives larger and high low ppm values. On substitution, the electron density of C of  $\text{me}=\text{C}-\text{NH}_2$  and  $>\text{N}-\text{CO}-\text{N}(\text{CH}_3)-$  increases upon addition of  $\text{CH}_2(\text{OH})_2$  in place of H of the  $-\text{NH}_2$  group. The signal at 167.4 ppm has been assigned to the carbons of the triazine nucleus whose amino groups are not substituted, while the signal at 166.3 ppm is attributed to the carbons of the nucleus whose neighboring amino groups are monosubstituted. A further upfield shift of 0.3 ppm is assigned to one more substitution in the same amino group. This proves that substitutions of the triazine nucleus by methylol shifts the band to a lower field. It is also observed in the case of methylurea. The signals in the region of 158.2 to 159.8 ppm are due to the carbonyl groups of methylurea residues. This is also found to give upfield on substitution, but in both cases the substitutions shift is found between 0.05 to 0.10 ppm. The signal at 71.4 ppm represents the carbons of methylol groups on the tertiary N atoms of the disubstituted amino groups on both the methylurea as well as the triazine ring. The signals at 67.6 and 68.6 ppm are assigned to methylate groups on the secondary N atom of methylurea and triazine, respectively, and confirms that the lower the substitution of the  $-\text{NH}_2$  group, the greater the upfield shift. The monosubstituted methylol groups on the secondary nitrogen atom of triazine and methylurea appear at 64.2 and 63.9 ppm, respectively. The methylene units in a branch appear at 54.1 ppm, while the methylene units chain have been assigned to the signals from 45.7 to 47.0 ppm.

Here, the thermal gravimetric analysis (TGA) represents weight change/loss of the resin, the thermal stability, and compositional analysis of samples. DTG kinetic, together with both curves, furnish an authentic information about degradation process of resin, and represent the thermograms; DTG highlight the curves of the resin and blends. The pure resin and

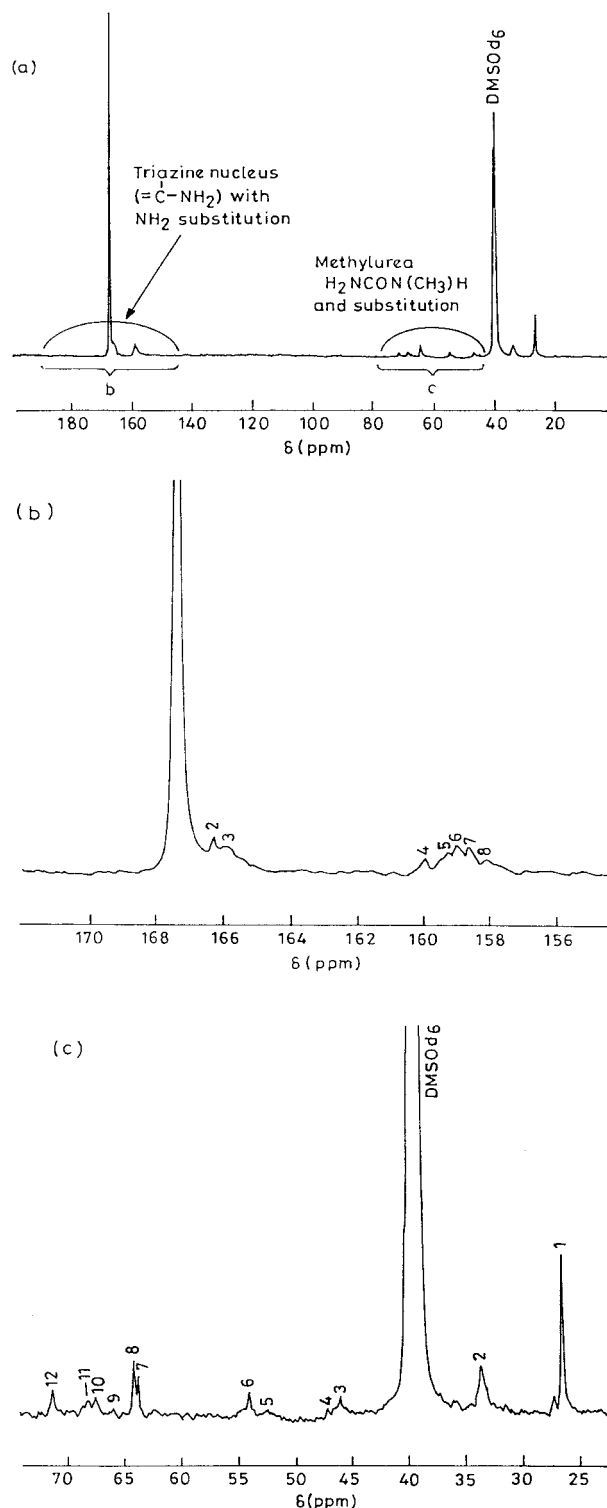
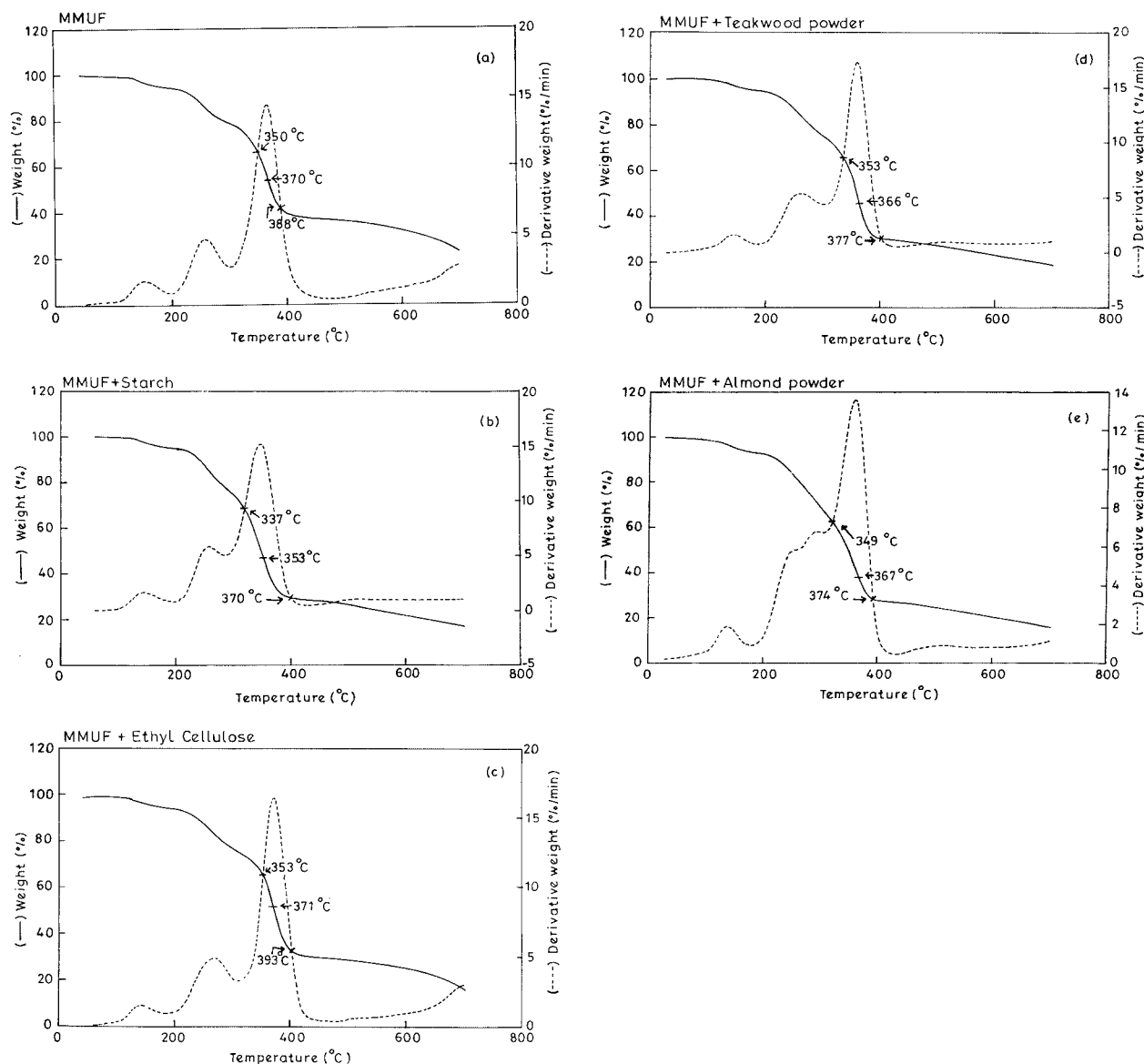


Figure 2 (a-c)  $^{13}\text{C}$  NMR, MMUF resin.

blends of ethyl cellulose are marked with four reaction zones, whereas the remaining blends find three major reaction areas. These are (1) from 120 to 190°C; this could specify that in this region a reaction of hydroxy methyl groups and dimethylene ether bridges with emission of water and formaldehyde undertakes. (2)



**Figure 3** TGA, (a) MMUF, (b) MMUF + starch, (c) MMUF + ethyl cellulose, (d) MMUF + teakwood powder, (e) MMUF + almond powder.

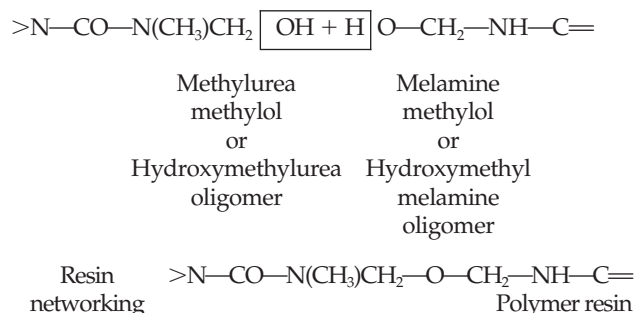
From 250 to 300°C, an oxidative degradation of methylurea units seems to occur. (3) From 350 to 400°C, i.e., an oxidative degradation of melamine units,<sup>48,49</sup> as the degradation process of resin and cellulosic blends by breaking the bonds release oxygen, which is used for oxidative degradation (4) demarcates the degradation of the residual fraction that starts from 450°C. It is evident that methylurea methylol undergoes an oxidative degradation at lower temperature than those of the melamine ones. Still, the residual fractions further undergo degradation after 450°C, which proves that teakwood, almond shell powder, and cellulose blends degrade at lower temperature than the resin and blends of ethyl cellulose. Therefore, ethyl cellulose could be applied for developing the blends of high thermal stability. After 450°C it seems that the resin network undergo degradation.

In the first range of 120 to 190°C, the temperature where the first-major loss in weight of the blends occurs is at about 140°C. This proves that the blending materials do affect the polymer matrix approximately to an equal extent, and the second major loss occurs at a similar temperature for the blends except almond shell powder, which shows two losses. This refers to the fact that the blending materials have little or almost a nil effect on the polymer matrix in these regions of temperatures. The third major loss of weight takes place at lower temperatures in the case of blends than that of the resin. The temperatures in the case of starch and the almond shell blended resins are found at 370 and 374°C, which are almost same, and indicates that both the starch and almond powder may have swelled out inside the polymer matrix, causing its breakage at lower temperature. It is also observed

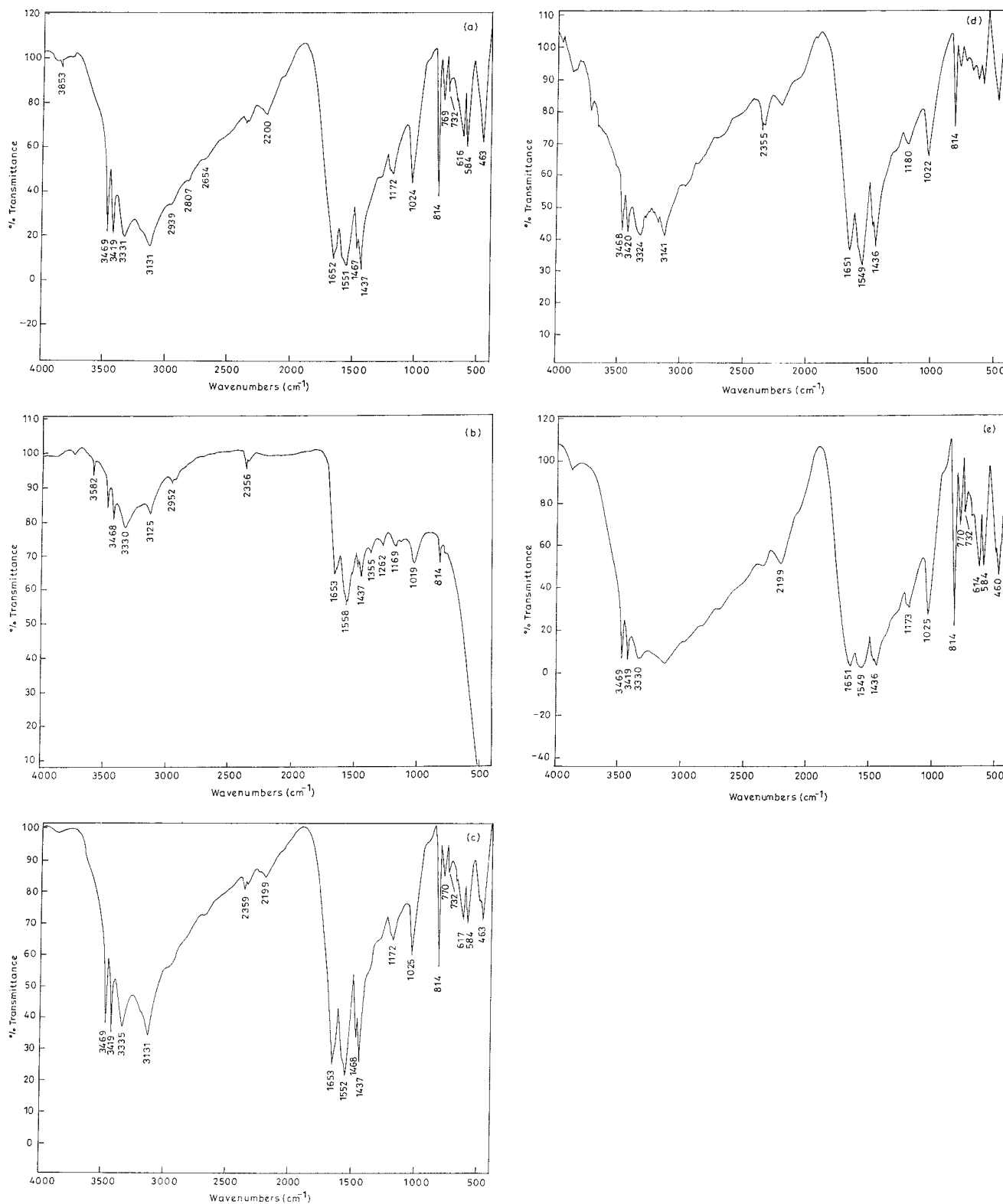
that the resin and ethyl cellulose lose 80% weight at 700°C, while the remaining blends do lose the same weight in the range of 350 to 400°C. This could be attributed to a degradation process of the resin or of the cellulose by bond breaking that generates oxygen that enhance the degradation process. Thus, interestingly, the blending materials accelerate the thermal degradation of the resin, and this mechanism is the required polymeric materials.

As is evident, the polyfunctional binding sites, due to a larger number of substitutionable  $\text{—NH}_2$  of melamine and methylurea, seems responsible for a variety of IR bands. The methylol,  $\text{CH}_2(\text{OH})_2$ , a binder after the oligomer formation for the reagents, does cause further functional sites, and finally, oligomers for resin network. Therefore, the several sites that affect IR spectroscopy do result in a complex spectra. Also, the blends do affect them, and hence, several stretching and bending vibration frequencies are reported in Figure 4 (a)–(e) for the resin and blends. The band at about  $1652\text{ cm}^{-1}$  is found due to the stretching of  $\geq\text{C=O}$  of the amide group of methyl urea ( $\text{—CONHCH}_3$ ) and  $\text{>C=N}$  stretching vibrations of the triazine ring. The band centered at  $3330\text{ cm}^{-1}$  consists of a superimposition of stretching vibrations of  $\text{—OH}$  groups with the  $\text{>NH}$  stretching vibrations. The bands at about  $3470$  and  $3419\text{ cm}^{-1}$  are due to  $\text{>N—H}$  stretching vibrations of primary amino groups of the triazine ring. The bands at about  $3330$  and  $3130\text{ cm}^{-1}$  are a superimposition of absorption due to  $\text{>NH}$  stretching vibrations of amino groups of amide and triazine units. These four bands indicate the presence of free amino groups in polymer and blends. The shoulder at  $3225\text{ cm}^{-1}$  is assigned to the  $\text{>N—H}$  stretch of bonded  $\text{>NH}$  in amide as well as triazine; the band at  $3130\text{ cm}^{-1}$  is attributed to substitution in the amino groups of amide. There seems to have been a combination of vibrations in various phases of  $\text{C—N}$  and  $\text{—NH}_2$  of amide and melamine, which appear at about  $1560\text{ cm}^{-1}$ , and also, the band at  $1440\text{ cm}^{-1}$  characterizes the stretching vibrations of amide as well as the triazine unit. The melamine does have a heteroatomic aromatic ring that is depicted by the bands at  $815$  and  $770\text{ cm}^{-1}$ , the oligomers of which are condensation products of methylurea and melamine with  $\text{CH}_2(\text{OH})_2$  indicated by a band at  $1021\text{ cm}^{-1}$ . Also, the band at about  $1140\text{ cm}^{-1}$  witnesses the crosslinking to some extent in the resin, which seems to be a function of temperature and exposure time of reaction mixture at any desired temperature of  $65^\circ\text{C}$ . It is observed that the IR spectra of the resin and blends are almost similar. However, the band at about  $3350\text{ cm}^{-1}$  is different in resin and blends. This explains the extent of blending on the basis of physical interaction of the blending material with the polymer resin through hydrogen bonding due to the presence of methylol groups and amino groups. The width of a

band at  $3330\text{ cm}^{-1}$  is almost similar for the resin and teakwood blend, but it is slightly broader for the ethyl cellulose. Further, it is much broader for the starch blend, this does depict an increase in interaction [Fig. 4(b)] of the polymer matrix with free  $\text{—OH}$  groups of starch and methylol of resin. This infers a greater possibility of linkages of starch with resin network or oligomers. The broadness of the band is reported due to hydrogen bonding.<sup>50,51</sup> The chosen polysaccharide blending materials are used as probe to investigate an effect on the structure and morphology of the resin, and in lieu the physicochemical properties of the resin blends. The  $^{13}\text{C}$  NMR and IR observations could be concluded with a suitable structure of oligomers and their networking as polycondensation of methylurea and melamine oligomers to form resin monomer is as



The scanning electron microscopy (SEM) is of interest for the bulk structure of the composites or blends for the homogenous distribution of their individual phases in each other. Figure 5(a)–(e) depict the SEM of the MMUF and blends. The micrograph of unblended resin is found uniform with respect to the distribution of the polymer matrix, and of the ethyl cellulose and starch, are alike, showing a dispersion of particle inside the polymer matrix including several types of patches. The almond and teakwood powder blends indicate the crack marks in the polymeric matrix system perhaps due to an integrated unit of oligomers. Thus, the micrographs support the thermal studies because almond shell powder is deeply penetrated or embedded in the polymer matrix. Probably it might considerably decrease the thermal decomposition of the blends, whereas in the case of the blend of starch the blown white spots on the micrograph are seen. This may be attributed to the swelling of starch with an increase in temperature. That reduces the thermal decomposition of the blend. This fact of starch blend further supports the broadening of the IR band of MMUF with starch as swelling is associated with the penetration of water molecules through hydrogen bonding. Comparatively, the blends of starch and ethyl cellulose find reasonable similarity in micrographs, which resembles the SEM micrograph of pure MMUF while the teakwood and almond powders do show the matching in crack marks; however, more marks are observed with the almond blend. It could be

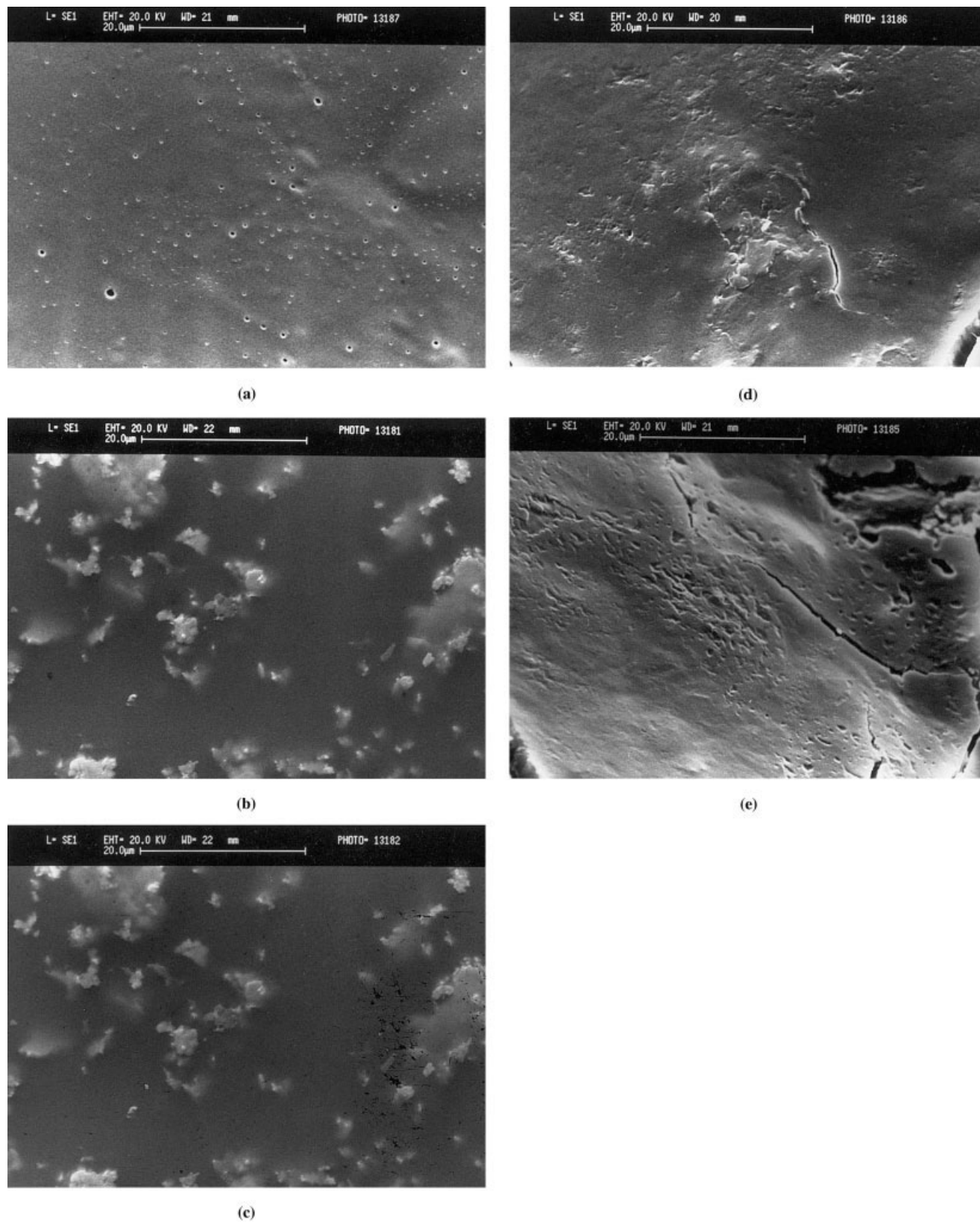


**Figure 4** FTIR, (a) MMUF, (b) MMUF + starch, (c) MMUF + ethyl cellulose, (d) MMUF + teakwood powder, (e) MMUF + almond powder.

shown that the starch and ethyl cellulose seem to establish an interaction with the resin matrix while teakwood and almond are weaker in this respect. This

presence of lignin and hemicellulose in teakwood powder and lignocellulose in the almond shell powder do introduce such behavior in the resin.





**Figure 5** SEM, (a) MMUF, (b) MMUF + starch, (c) MMUF + ethyl cellulose, (d) MMUF + teakwood powder, (e) MMUF + almond powder.

## CONCLUSION

Thermal studies indicate that the blending with starch, teakwood, and almond shell powder reduces the thermal degradation of the resin. The results of IR studies as well as by Scanning Electron Micrograph support the degradation trend. Thus, the blending may prove to be ecofriendly in applications, thus broadening the scope of utility. This information could be of some significance regarding the resins of di- and trimethyl N-derivatives of urea.

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