



HARMONIZING STRENGTH AND SUSTAINABILITY: EMA-Na ENHANCED LOW-DENSITY POLYETHYLENE-STARCH BLENDS

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Abstract

Combinations of low density polyethylene and starch, compatible with maleic anhydride (MA) and the sodium salt of polyethylene-co-methacrylic acid (EMA-Na), were made. By analysing mechanical characteristics, melt flow measurements, spectroscopy, thermal analysis, and scanning electron microscopy, the compatibility behaviour of blends was investigated. mixes without compatibilizer exhibit the undesirable characteristics of polymer mixes that are incompatible. The ionomer is an effective compatibilizer for low density polyethylene-starch blends, according to both spectroscopic and morphological characteristics. Based on biodegradability testing, the blends show some degree of biodegradability.

Keywords: compatibilizer, low density polyethylene, starch, ionomer, biodegradability

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INTRODUCTION

The low cost, simplicity of processing, insensitivity to moisture, and flexibility of low density polyethylene (LDPE) make it a popular choice for packaging. Nevertheless, LDPE is not naturally biodegradable.¹ Relatively cheap biopolymers with renewability and biodegradability are starch and other biopolymers.^{2,3} A tiny quantity of starch added to low density polyethylene can improve the blends' ability to decompose naturally. In order to create new materials with specific qualities, mixing low density polyethylene with biopolymers presents an intriguing option.

The majority of polymer blends are determined to be incompatible and immiscible. The general physico-mechanical properties of miscible blends are determined by two structural parameters: (a) appropriate interfacial tension, which results in a phase size small enough to permit the material to be regarded as macroscopically homogenous, and (b) an interphase adhesion strong enough to absorb stresses and strains without causing the material's established morphology to be disrupted.^{4,5}

Large particles in the dispersed phase are the result of weak interfacial adhesion and high interfacial tension in immiscible blends formed by starch and low density polyethylene. The weak stress concentration at the polymer-polymer interface of these immiscible blends prevents the tension from moving from the continuous phase to the dispersed phase while they are under stress. Poor mechanical qualities are the outcome of this incompatibility in blends.⁶ Numerous studies have mentioned the use of compatibilization to enhance the qualities of the blend, ranging from altering one of the blend components^{7,8} to adding a minor-component compatibilizer.⁹⁻¹² A compatibilizer is added to the blend to improve its mechanical characteristics by

increasing the stress transmission between the dispersed and continuous phases.¹³

Numerous research reports have been released about the use of compatibilizers to enhance the morphological and mechanical properties of LDPE blends. It has been found that grafted polyethylene with maleic anhydride functions as a compatibilizer in LDPE blends.¹⁴ In LDPE blends, grafted LDPE with glycidyl methacrylate has also been employed as a compatibilizer.^{15,16}

A hydrocarbon backbone with attached acid groups that are at least partially neutralised to create salt groups is what makes an ionomer, an ionic polymer.^{17,18} Growing interest in industry and academia is sparked by the special qualities of ionomers. Within contemporary polymer science, one of the most active fields of research is the study of ionic interaction in macromolecular systems. The special property of ionomers allows them to reconcile blends that would otherwise be incompatible, such as PP/EVOH¹⁹, LDPE/Nylon-6^{20,21}, HDPE/Nylon-66²², and so on. Several ionomers have been effectively employed in the literature to compatibilize incompatible blends, including metal salts of sulfonated PET²³, poly(ethylene-co-sodium methacrylate)²⁰, and poly(styrene-co-sodium methacrylate).²⁴ At the interface between the blends, ionomers that are introduced to the binary mix create ionic cross-links and thus homogeneity is reinforced.

The investigations on the usage of maleic anhydride (MA) and ionomer (the sodium salt of polyethylene-co-methacrylic acid, or EMA-Na) as compatibilizers for low density polyethylene and starch blends are presented in this paper. This paper uses maleic anhydride as a reference, as it has been frequently used as a compatibilizing agent for these kinds of systems. Table 1 provides the names and descriptions of the samples that were used.

Table 1. Description of sample designations

<i>Sample designation</i>	<i>Description</i>
C5M	LDPE-20% starch-5% MA
C5S	LDPE-20% starch-5% (EMA-Na)
LDS(x)-MA(y)	LDPE- x% starch-y% MA
LDS(x)-Na(y)	LDPE- x% starch-y% (EMA-Na)

MATERIALS

Low density polyethylene (LDPE)

The film grade low density polyethylene (LDPE 24FS040) from Reliance Industries Limited, Mumbai, India, with melt flow index (190 °C/2.16 kg) of 4 g/10 min and density (23 °C) of 0.922 g/cm³ was supplied by Periyar Polyfilms, Edayar, Kerala, India.

Starch

The tapioca starch (100 and 300 mesh) was obtained from Jemsons Starch & Derivatives, Aroor, Alappuzha, Kerala. As these fillers were hygroscopic in nature they were oven dried at 120 °C for 1h prior to mixing.

Ionomer

Ionomer used in this study was Sodium salt of poly(ethylene-co-methacrylic acid) (HIMILAN

1702 EMAANa) with melt flow index (190 °C/2.16 kg) of 10 g/10 min.

Ionomer was supplied by Mitsubishi Plastics, Inc., Japan.

METHODS

Preparation of blends

A Thermo Haake PolyLab system (Rheocord 600p) equipped with roller-type rotors was used for melt mixing. The mixing chamber has a volumetric capacity of 69 cm³. A mixing time of 8 minutes was given for all the compounds at a rotor speed of 30 rpm at 150 °C. LDPE together with ionomer was first melted for 2 minutes followed by the addition of filler. Mixing was continued for another 6 minutes.

Preparation of test specimens

The test specimens were prepared from neat LDPE and the compounds by moulding in an electrically heated hydraulic press for 5 minutes at 150 °C under a pressure of 20 MPa. After moulding, the samples were cooled down to room temperature under pressure.

Characterization

Mechanical properties

The mechanical properties were evaluated using Shimadzu Autograph AG-I series universal testing machine at a crosshead speed of 50 mm/min. Tensile strength, elongation at break and elastic modulus were measured according to ASTM D-882 (2002). Averages of at least five sample measurements were taken to represent each data point.

Melt Flow Index (MFI)

The melt flow index (MFI) of each blend of LDPE with filler was measured using a CEAST Modular Line Melt Flow Indexer in accordance with ASTM method D-1238 using a 2.16 kg load at a melt temperature of 190 °C.

Biodegradation studies

The biodegradation studies on the blends were carried out according to ASTM D-6691. Bacterial cultures were obtained from culture collections of *Microbial Genetic Lab, Department of Biotechnology, Cochin University of Science and Technology*. These cultures were isolated from sediment samples collected from different locations in Cochin backwaters and Mangalavanam mangroves. These cultures were previously identified as the genus *Vibrionacea* based on their morphological and biochemical characteristics outlined in Bergey's Manual of Systematic

Bacteriology.¹⁵ They were preserved in 10mL glass bottles employing the paraffin oil overlay method. To prepare the inoculum the individual isolates of the consortium were grown overnight at 37 °C at 120 rpm on an Orbitek shaker (Scigenics Pvt. Ltd, Chennai, India) in nutrient broth (Himedia, Mumbai) pH 7.0 ± 0.3 with 1% NaCl. The cells were harvested by centrifugation at 5000 rpm (2292 g) for 20 minutes, washed with physiological saline and then pooled. 5mL of this pooled culture (OD₆₆₀ = 1) was used to inoculate 50mL amylase minimal medium¹⁷ lacking starch. The samples prepared from the blends previously wiped with 70% alcohol were added to this medium and these strips acted as the sole source of carbon. Incubation was in the Orbitek environmental shaker at 37 °C and 120 rpm for a total period of 3 months with regular sampling. The medium without the inoculum with corresponding starch-plastic blends and subjected to the same treatment as above were used as controls.

Water absorption characteristics

Water absorption was measured using 3 x 1 inch film strips of <1mm thickness according to ASTM D-570-81 method. Water absorption measurements were performed by soaking the samples in distilled water. The water absorption was calculated as the weight difference and is reported as a percentage increase of the initial weight. The results reported are average of three measurements.

Fourier transform infrared spectroscopy (FTIR)

The FTIR spectra of the samples were recorded in the transmittance mode using a Thermo Nicolet, Avatar 370 FTIR spectrophotometer in the spectral range of 4000–400 cm⁻¹.

Morphological studies

In the present study the tensile fractured surfaces were mounted on a metallic stub with the help of a silver tape and conducting paint in the upright position and were sputter coated with platinum within 24 hours of fractures in a JFC 1600 Autofine coater and then examined under JEOL model JSM-6390LV scanning electron microscope (SEM).

RESULTS AND DISCUSSIONS

The mechanical properties of the compatibilized as well as uncompatibilized LDPE-starch blends are shown in the figures 1, 2 and 3. Because hydrophilic starch and hydrophobic LDPE have weaker interfacial adhesion, the tensile strength dropped as the concentration of starch increased.^{25,26} The tensile strength of samples

containing EMA-Na and maleic anhydride as compatibilizing agents is higher than that of blends that are not compatible. The maximum tensile strength for blends that are compatible with MA is obtained by adding 2% of MA, but for blends that are compatible with ionomers, the maximum tensile strength is obtained by adding 5% of

ionomer in the case of EMA-Na. It appears that the variations in phase behaviour are what cause the fluctuations in tensile strength. Compatibility between LDPE and starch rises as a result of compatibilizer addition, which also increases interfacial adhesion and promotes effective stress transmission from one phase to the next.

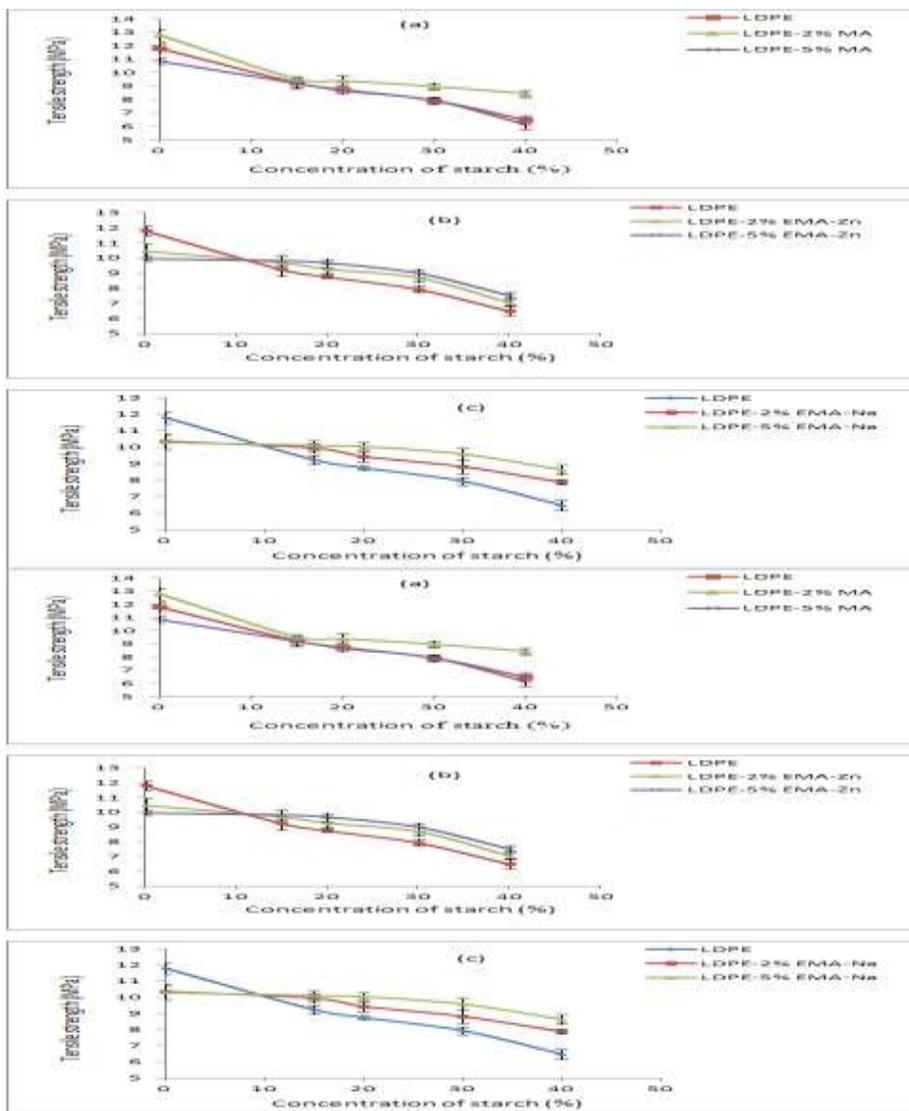


Figure 1. Variation of tensile strength with the concentration of starch for: (a) LDPE-starch -MA blends and (c) LDPE-starch-(EMA-Na) blends

The ductility, as measured by the elongation at break of the blends, is shown in Fig 3.2. In comparison to the uncompatibilized blend, the elongation at break increased for all compatibilized

blends. The increased ductility indicates compatibilization has occurred and is ascribed to the higher interfacial adhesion and the decrease in particle size.

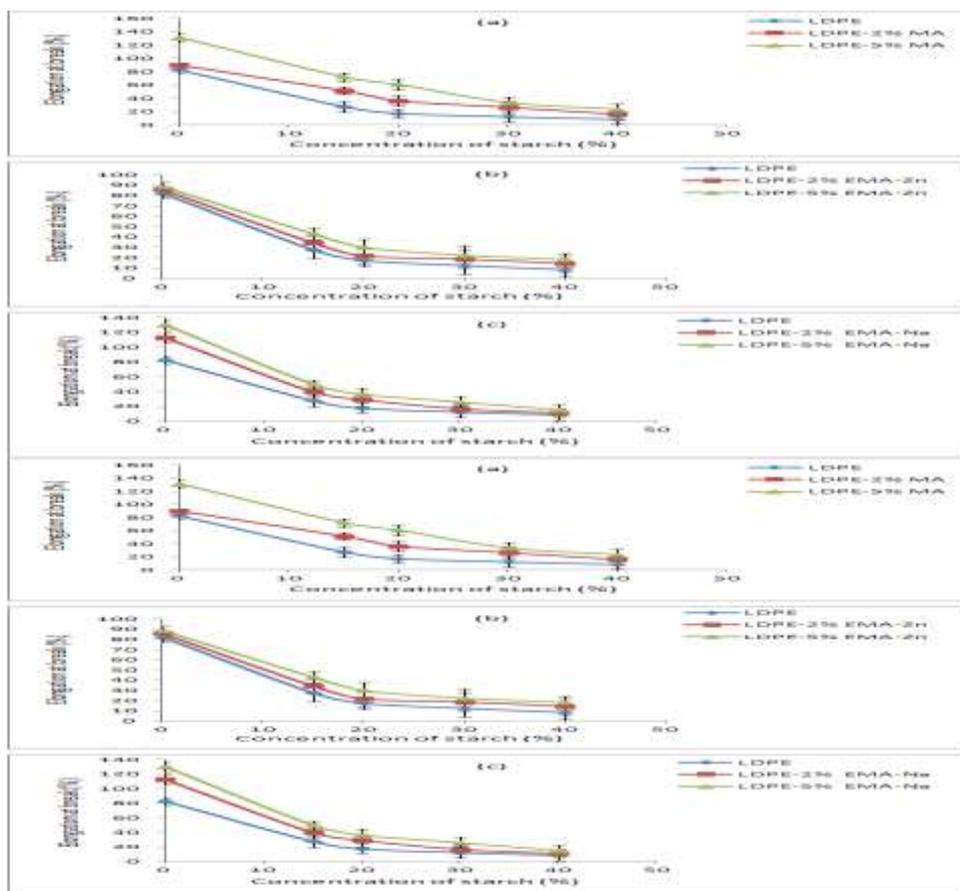
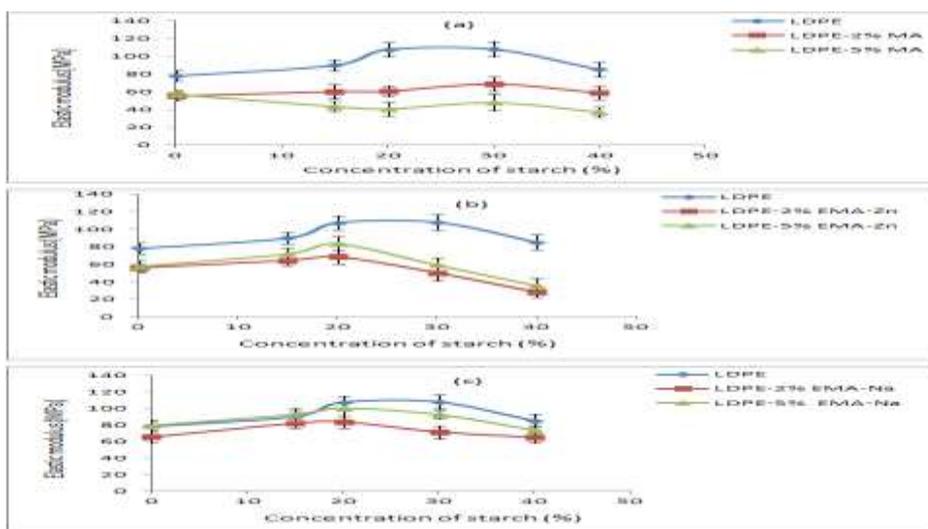


Figure 2. Variation of elongation at break with the concentration of starch for: (a) LDPE-starch -MA blends and (c) LDPE-starch-(EMA-Na) blends

Conversely, as the starch loading rises, the elastic modulus rises as well (figure 3.3). After processing, starch integrated into LDPE seems to hold onto its granular form. Because they are hard, these grains serve as stiff fillers. Generally speaking, modulus and the material's hard domain are tightly

associated. The hard domain content and the blend's tensile modulus both rise with the amount of starch present. However, because of the blends' flexibilization, the compatibilized blends' elastic moduli are lower than those of the uncompatibilized blends.



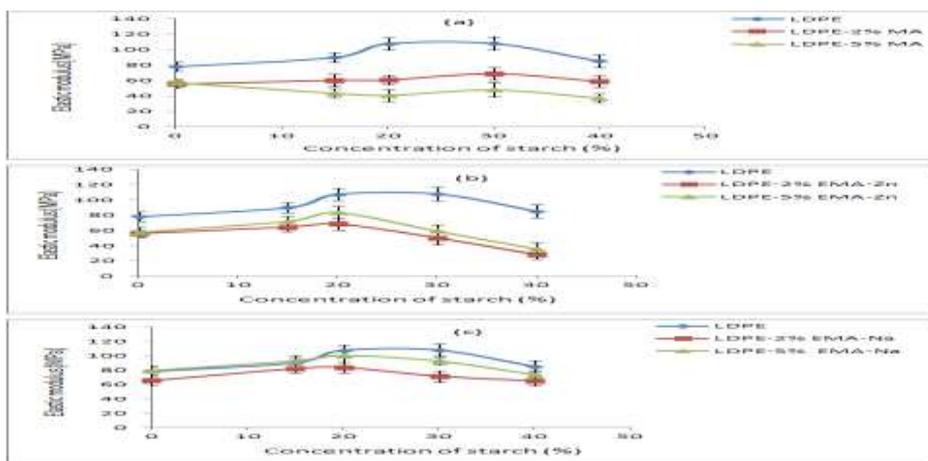


Figure 3. Variation of elastic modulus with the concentration of starch for: (a) LDPE-starch -MA blends and (c) LDPE-starch-(EMA-Na) blends

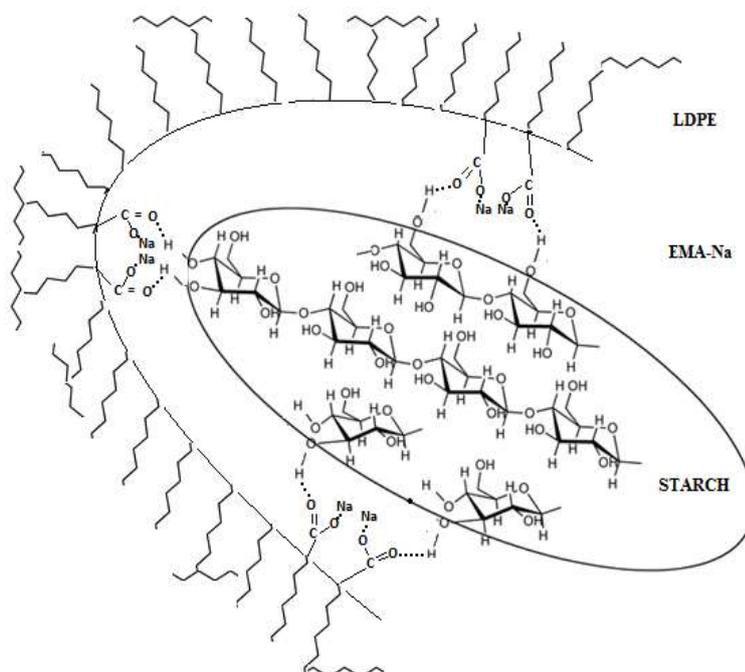


Figure 4. Proposed schematic representation of the interaction between starch, EMA-Na and LDPE

Homogeneity appears to be enhanced when ionomers are introduced to the binary mix because they appear to produce ionic cross-links at the blend interface. A polyethylene-rich domain and a region primarily made up of a metal cation-carboxylate anion pair are the two unique areas of the ionomers used in this investigation. It is possible that the hydroxyl groups of starch and the carboxyl groups of the ionomers are engaging through polar-polar interactions, as Figure 4. suggests. The ionomers' nonpolar polyethylene domain and the LDPE's are compatible. It is thought that the LDPE and the ionomers' polyethylene domains associate via co-crystallization, amorphous chain entanglement, or both.^{27,28} Although there is a modest van der Waals type connection between the nonpolar LDPE molecular chains and the nonpolar region of the ionomers, there may be a much stronger type

interaction because of the hydroxyl groups in starch and the carboxyl groups in the ionomer, as shown in the images.

Up to 5% of the weight is added in ionomer content, which results in a tensile strength improvement. More than five weight percent of ionomer does not lead to any additional tensile strength enhancement. This implies that the strength of the matrix, which is greatly influenced by the amount of compatibilizer in the blend, as well as interfacial adhesion determine the tensile strength of a compatibilized blend.²⁹ The reason for the lack of improvement in characteristics at higher ionomer concentrations in the blend could be attributed to either the ionomer's restricted solubility in the blend or the migration of ionomer to the interface during processing being limited.³⁰

The effect of the addition of the ionomer as compatibilizer on the tensile strength of LDPE-starch blends is shown in Figure 5.

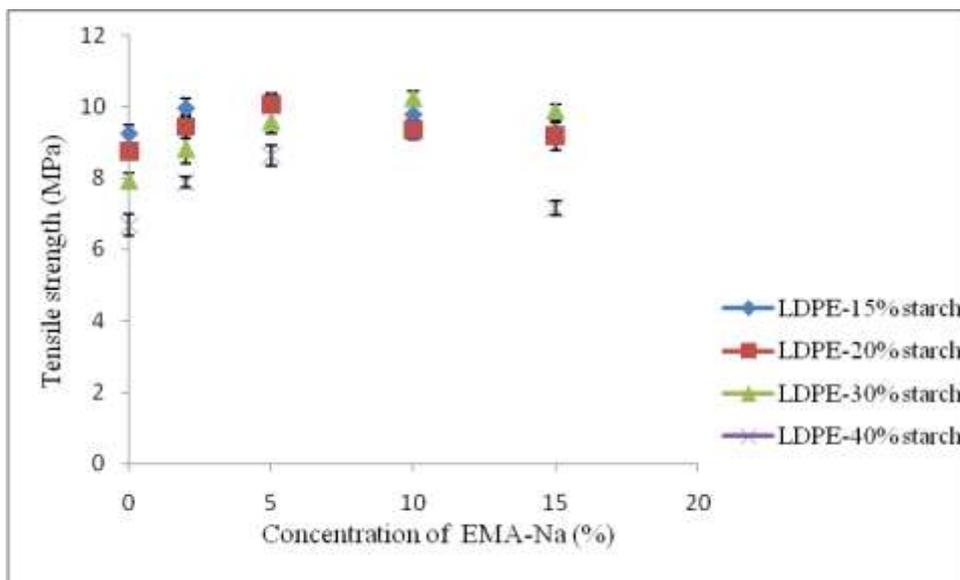


Figure 5. Effect of concentration of EMA-Na as compatibilizer on the tensile strength of LDPE- starch blends

Melt flow measurements

Figure 6. show the variations of melt flow indices with varying concentration of starch in the case of LDPE-starch-(EMA-Na) blends. The inverse of melt viscosity, the melt flow index is a measure of average molecular mass. When compared to samples without starch, the MFI values of all the samples with starch are lower. As the starch concentration rose, the MFI values dropped. This might be the result of an increase in viscosity brought on by a concentration of spherical starch

particles in the LDPE matrix. It was shown that when the ionomer content rose from 2 weight percent to 15 weight percent, the melt flow increased in all cases of LDPE-starch-ionomer blends. Despite the fact that ionomers often exhibit higher melt viscosities than their base polymers, the ionomers employed in this work may have had an increase in melt flow because of their low molecular weight backbone.³¹ This demonstrates unequivocally the beneficial compatibilizing action of ionomer.

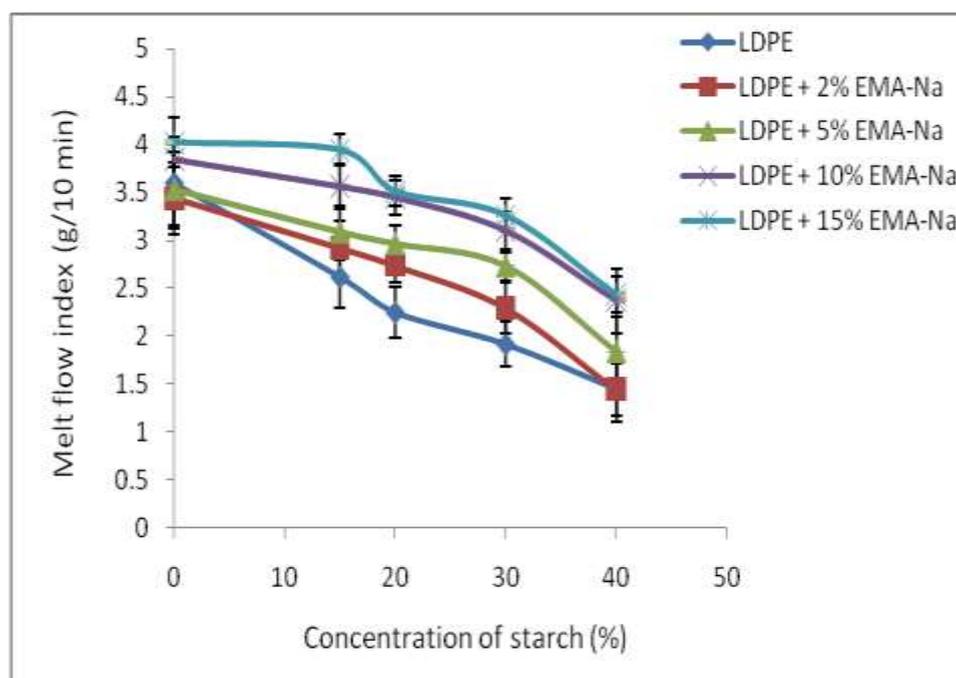


Figure 6. Variation of melt flow index with the concentration of starch in LDPE-starch-(EMA-Na) blends

Biodegradation studies

Figure 7. exhibits the tensile properties of LDPE-20% starch-compatibilizer blends after biodegradation in culture medium. Following biodegradation, all blends exhibit a notable decrease in tensile strength. The consumption of starch by microbes is the cause of this decrease. The bigger decline in stress-strain characteristics following biodegradation in the blends containing higher starch content suggests that the starch in these blends is more exposed and, consequently, more of it is eaten by microorganisms. Microorganisms may not be able to access starch in blends with lower starch content because the starch may be nearly entirely covered in LDPE.³²

The tensile properties of the compatibilized blend films were higher than those of the uncompatibilized blend films during a four-month biodegradation period in culture medium. It appears that in the uncompatibilized blends, the starch granules are encapsulated by the LDPE matrix without any bonding. This leads to the LDPE matrix's porosity, which facilitates microbial invasion. The removal of starch granules from the LDPE-starch film with compatibilizer may be more challenging due to the interfacial adhesion between the two components.

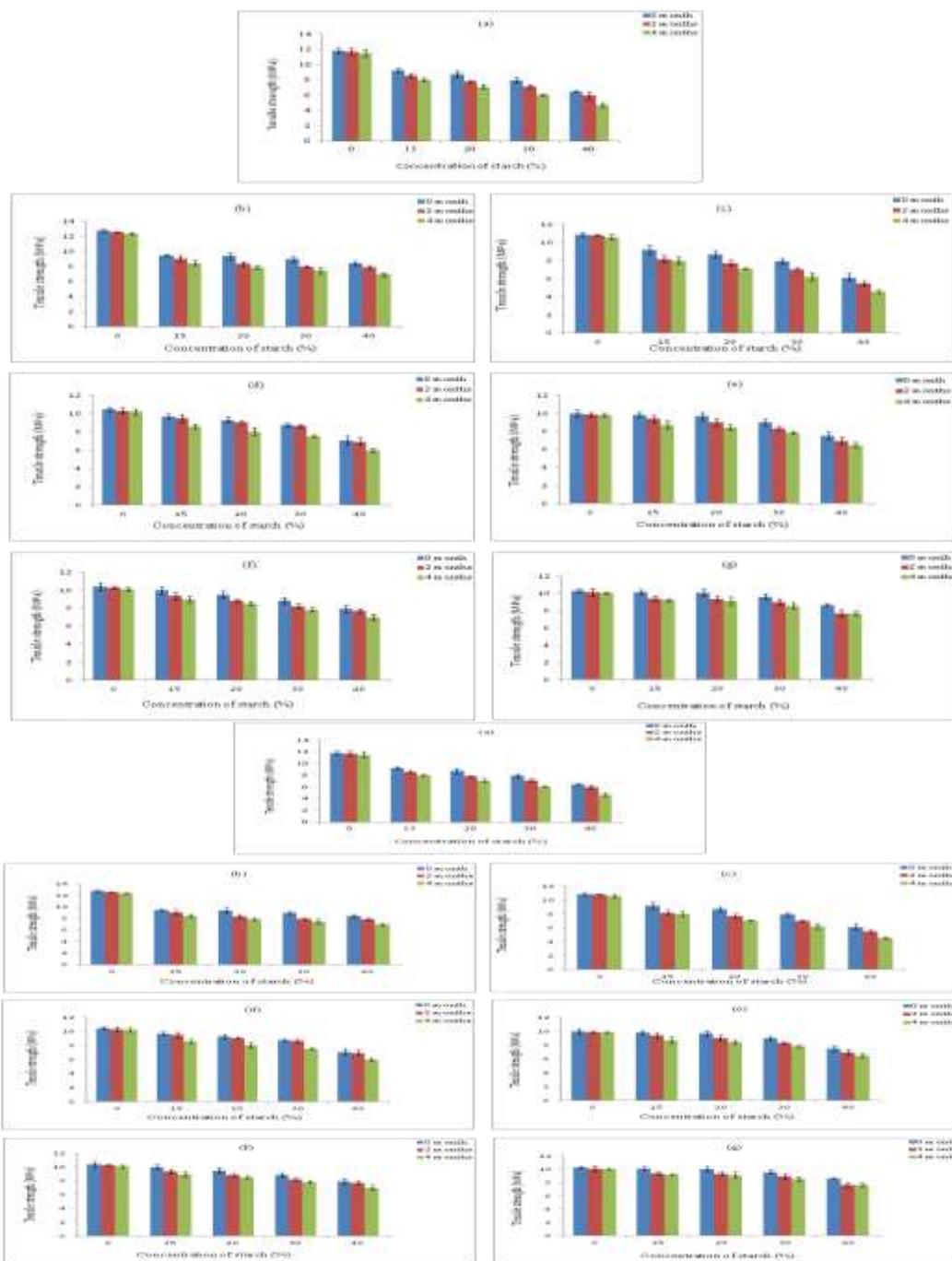


Figure 7. Biodegradation of: (a) LDPE-starch blends, (b) LDPE-starch-2% MA blends, (c) LDPE-starch-5 % MA blends, (f) LDPE-starch-2% (EMA-Na) blends and (g) LDPE-starch-5% (EMA-Na) blends

Tables 2 and 3 illustrate the weight loss of compatibilized LDPE-starch blends after biodegradation in culture medium for four months.

Table 2. Percentage decrease in weight of LDPE-starch-MA blends after biodegradation in culture medium for four months

<i>Sample</i>	<i>Initial weight (g)</i>	<i>Weight after four months (g)</i>	<i>% weight loss</i>
LDS(0)-MA(2)	0.3042	0.3041	0.03
LDS(0)-MA(5)	0.2254	0.2253	0.04
LDS(15)-MA(2)	0.2411	0.2320	3.77
LDS(15)-MA(5)	0.2769	0.2659	3.97
LDS(20)-MA(2)	0.2155	0.2056	4.59
LDS(20)-MA(5)	0.2858	0.2669	6.61
LDS(30)-MA(2)	0.1888	0.1718	9.00
LDS(30)-MA(5)	0.1583	0.1409	10.99
LDS(40)-MA(2)	0.3141	0.2445	22.16
LDS(40)-MA(5)	0.1552	0.1095	29.45

Table 3. Percentage decrease in weight of LDPE-starch-(EMA-Na) blends after biodegradation in culture medium for four months

<i>Sample</i>	<i>Initial weight (g)</i>	<i>Weight after four months (g)</i>	<i>% weight loss</i>
LDS(0)-Na(2)	0.2688	0.2687	0.04
LDS(0)-Na(5)	0.3144	0.3143	0.03
LDS(15)-Na(2)	0.2665	0.2601	2.40
LDS(15)-Na(5)	0.1486	0.1466	1.35
LDS(20)-Na(2)	0.2168	0.2074	4.34
LDS(20)-Na(5)	0.2969	0.2874	3.20
LDS(30)-Na(2)	0.2154	0.2001	7.10
LDS(30)-Na(5)	0.2761	0.2604	5.69
LDS(40)-Na(2)	0.2092	0.1794	14.25
LDS(40)-Na(5)	0.1902	0.1696	10.83

The weight loss observed in all compositions is attributed to the microbes' consumption of starch. While blends of LDPE with starch showed higher weight reduction, LDPE alone did not demonstrate any discernible weight loss in culture medium. For the mix containing 40% starch, it was shown that the biodegradation rate increased quickly. As shown in the table, the MA compatibilized films

had a higher degradation rate than the ionomer compatibilized films. Possible implications include the compatibilizer's potential to prevent the film from biodegrading. The polar-polar interactions that prevent bacteria from consuming starch may be the cause of this ionomer effect. These interactions include the carboxyl groups of the ionomer and the hydroxyl groups of starch.

Water absorption studies

Table 4. Water absorption of LDPE-starch-(EMA-Na) blends

<i>Sample</i>	<i>Initial weight (g)</i>	<i>Weight after 24 hours (g)</i>	<i>% water absorption</i>
LDS(0)-Na(2)	0.3682	0.3684	0.05
LDS(0)-Na(5)	0.3773	0.3774	0.03
LDS(15)-Na(2)	0.3021	0.3043	0.73
LDS(15)-Na(5)	0.5418	0.5450	0.59
LDS(20)-Na(2)	0.3436	0.3480	1.28
LDS(20)-Na(5)	0.5418	0.5485	1.25
LDS(30)-Na(2)	0.4653	0.4723	1.50

LDS(30)-Na(5)	0.3344	0.3389	1.35
LDS(40)-Na(2)	0.2395	0.2473	3.26
LDS(40)-Na(5)	0.3513	0.3579	1.88

Table 4. shows the water uptake of LDPE-starch-ionomer blends after 24 hours of immersion. Since starch absorbs water at a far higher rate than LDPE, blends' water uptake rises as the starch content does.³³ On the other hand, ionomer addition reduces water absorption, with less water

absorption noted at larger ionomer levels. This might be the result of improved blend component adhesion brought about by the decrease in void volume in ionomer compatibilized blends, which limits water storage and penetration at the interface.

Fourier transform infrared spectroscopic analysis

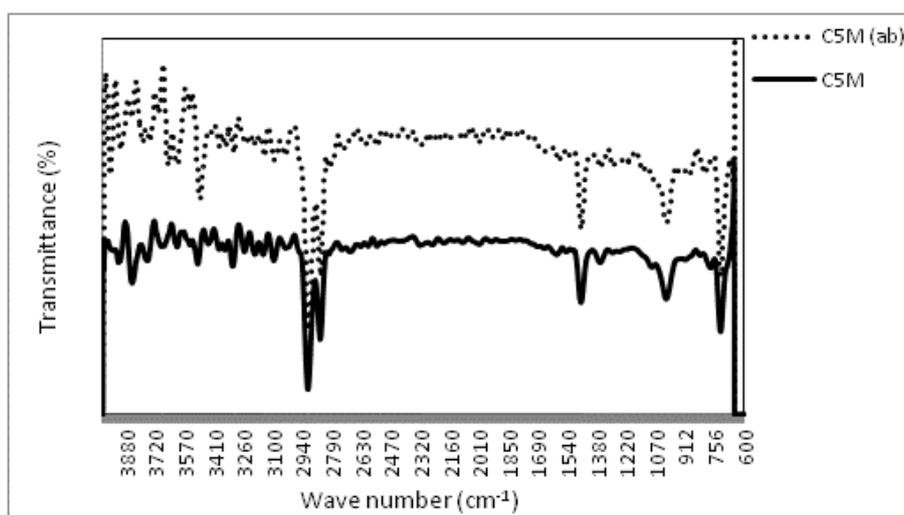


Figure 8. FTIR spectra of LDPE-starch-MA blends before and after (ab) biodegradation

The FTIR spectra of LDPE-starch-compatibilizer blends before and after biodegradation for four months are shown in figures 8 and 9. After deterioration, the peaks at 2921–2848 cm^{-1} , 1473–1463 cm^{-1} , 1156–1028 cm^{-1} , and 730–720 cm^{-1} showed increased intensities. This could be because the polyethylene chain broke in degradable

settings, leading to an increase in the quantities of terminal groups. The C-O stretching absorbance at the 1260–1000 cm^{-1} region of the C5M blend's spectra before and after biodegradation differs noticeably, suggesting that the starch has been removed from the plastic film.

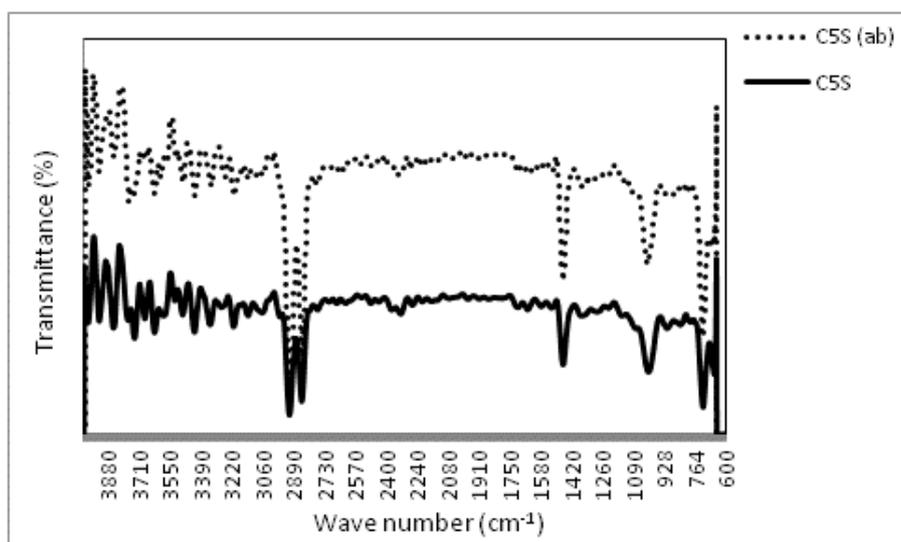


Figure 9. FTIR spectra of LDPE-starch-(EMA-Na) blends before and after (ab) biodegradation

This may be because starch and ionomer have stronger phase adhesion in C5Z and C5S blends,

where these variations are less noticeable. The O-H stretching peak at 3700–3000 cm^{-1} region and the

O-H bending peak at 1640 cm^{-1} , which were both somewhat less intense after biodegradation, were seen in the spectra of the C5M and C5Z blends, and

these changes indicated that water was lost as absorbed when microorganisms degraded the starch.

Table 5. Characteristic FTIR spectral peaks in C5M and C5S

Sample	Peak position (cm^{-1})	Characteristic group
C5M	2913, 2847	C-H stretching
	1790	C=O stretching
	1591	C=O stretching
	1463	CH ₂ scissor and asymmetric bending
	1361	C-H bending
	1011	O-C stretching
	916	O-H deformation
	722	CH ₂ rocking
	C5S	2913, 2848
1735		C=O stretching
1700, 1522		C=O stretching
1463		CH ₂ scissor and asymmetric bending
1364		C-H bending
1010		O-C stretching
911		O-H deformation
721		CH ₂ rocking

Morphological studies

The morphology of the LDPE-starch-compatibilizer (75/20/5) blends is depicted in the figures indicated by (i). Although the two phase morphology looks a lot like the uncompatibilized blend, there are some starch particles that were broken during the cryogenic fracture, and there is some plastic deformation at the interfaces that suggests there were more interactions between the two phases. By decreasing the size of the dispersed

phase and enhancing its dispersion into the LDPE matrix, the compatibilizer's inclusion has a noticeable impact on the morphology, as demonstrated by the figures. This shrinkage implies a shrinkage of the starch agglomerates' size. Thus, well-dispersed starch particles are produced by the addition of ionomers. It is evident that compatibilization and a reduction in interfacial tension has place since the particle size distribution is more uniform and the total size is less.

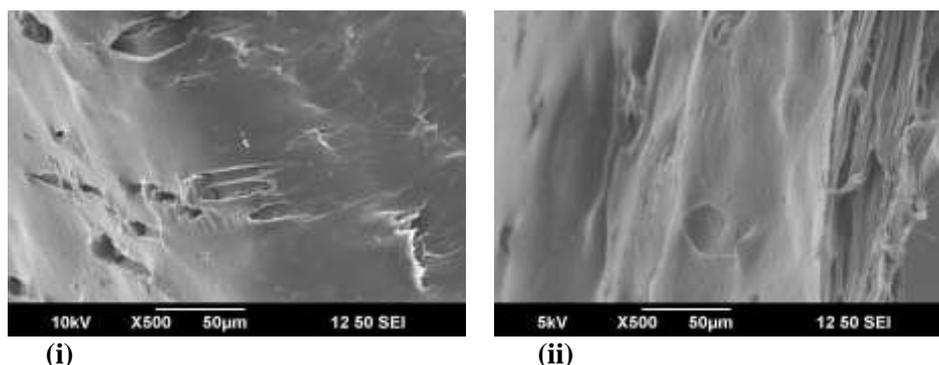


Figure 10. Scanning electron micrographs of LDPE-starch-MA (75/20/5) blend: (i) before biodegradation and (ii) after biodegradation for four months

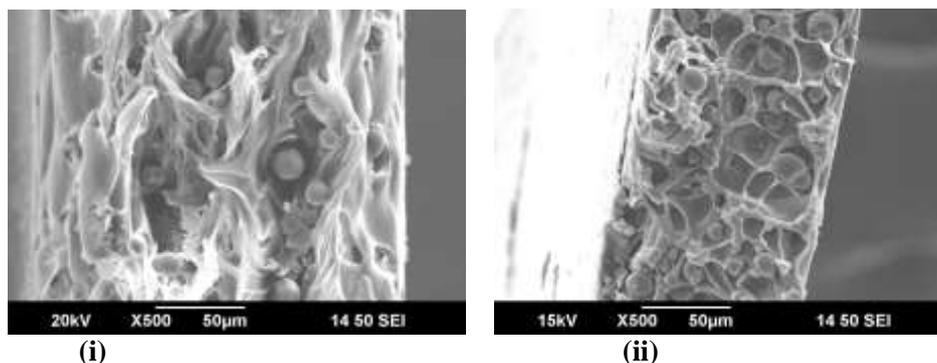


Figure 11. Scanning electron micrographs of LDPE-starch-(EMA-Na) (75/20/5) blend: (i) before biodegradation and (ii) after biodegradation for four months

Figures indicated by (ii) provide SEM micrographs of blends of LDPE, starch, and compatibilizer following biodegradation. The LDPE-starch blends' biodegradation is demonstrated by the micrographs' black pores. During the biodegradability tests, not all of the starch is eliminated. In comparison to the compatibilized film, the uncompatibilized film has higher surface area available for microbial attack, which leads to a greater number of sporadic tiny holes. The compatibilizer increases the interfacial adherence of starch into the LDPE matrix, which makes it more difficult to remove starch granules from the films.

CONCLUSIONS

The sodium salt of the polyethylene-co-methacrylic acid ionomer can be used to compatibilize melt-mixed blends of low density polyethylene and starch. The blends' stress-strain characteristics were enhanced by the compatibilizer (ionomer) addition. The study's ionomer dosages were varied, however for EMA-Na, the dosage of 5% demonstrated the greatest increase in mechanical qualities. The ionomer was added, and this resulted in an increase in the mixes' melt flow. Based on biodegradation investigations, it was shown that the ionomer compatibilized films had a somewhat slower rate of deterioration than the MA compatibilized films. Studies using spectroscopy reveal an interaction between the ionomer and starch. The presence of ionomers as compatibilizers in the mix improves the dispersion of starch particles, according to morphological investigations.

CRediT authorship contribution statement.

Author 1: Methodology, Investigation, Formal Analysis Author 2: Supervision.

Declaration of competing interest. The authors declare that we have no known competing financial interests or personal relationships that could have

appeared to influence the work reported in this paper.

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