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Tuning Performances of Poly(L-lactide-co-glycolide)s Through Microstructure and Glycolide Content for Sustainable Packaging Applications

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Research Article

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Abstract

Plastic plays an important role in the modern society due to its excellent properties and widespread applications. However, one of the biggest drawbacks of plastic is its nondegradability in the natural environment. Although many biodegradable materials have been reported, materials with adequate properties for sustainable packaging applications and disposable items are still lacked off. Herein, random and block poly(lactide-co-glycolide)s with different glycolide contents have been prepared in one-step reaction via ring-opening polymerization. The lactide-to-glycolide ratio was 1:1 and 1:9, respectively. Their potentials to replace traditional plastics have been fully evaluated as well. Through changing composition or microstructure, polymer properties can be controlled. All polymers shown high degree of crystallinity from 57-89 %, except **rPLGA5050** was amorphous. The increase of crystallinity leaded to enhancing thermal stability, mechanical properties, thermomechanical behaviours and permeability. The composition and microstructure also made significant contribution in manipulating degradation rate. Among polymers, **bPLGA5050 and bPLGA1090** exhibited superior properties compared to their random counterparts. The excellent balance between physical properties and degradability implied that they had strong potential for sustainable packaging applications.

Keywords: random copolymer; block copolymer; lactide; glycolide; poly(lactide-*co*-glycolide); polymer

1. Introduction

Global plastic production has sharply developed over the last 70 years due to enormous demand of human [1]. Plastic has added many values to our lives due to their versatile, low cost and sterility [2]. It is able to be used in various applications including medical instruments [3], construction [4], food packaging [5], sensing [6-8] and home appliances [9]. Annually, about 450 million tonnes of plastic are produced [10]. But this number also highlights a more insightful story, an equivalent amount of waste will be generated as well. About one-quarter of waste plastic is improperly treated or directly discarded into the ocean. Instead of being decomposed within a short interval, plastic waste lasts there for centuries [11]. Moreover, plastic can be also fragmented into microparticles, which are harmful to biodiversity, ecological system, economic activities and human health [12]. Nowadays, many countries have implemented plastic item bans [13]. From the facts, the development of new eco-friendly materials is essential.

To overcome the abovementioned issues, the use of degradable materials such as pectin [14], gelatine [15], starch [16], chitosan [17], polyglycolide [18], polylactide [19] and so on has gained significant attention more than ever before. Among them, polylactide and polyglycolide have emerged as promising candidates for widespread applications due to renewable origin, biodegradability and favourable mechanical properties [20, 21]. However, the poor barrier properties of polylactide compared to commonly used plastics significantly limit its ability in packaging application and disposable goods [21, 22]. Although polyglycolide exhibits excellent barrier properties, its inherent brittleness and high melting temperature pose a barrier for processing [23]. More importantly, the degradation of both polylactide and polyglycolide remains challenging as well. Polylactide often requires specific conditions for degradation and this process occurs at a relatively slow rate [24]. In the contrary, polyglycolide degrades too rapidly, which may also restrict its practical applicability [23]. Therefore, there is a need to find out new materials that can combine the desirable properties of both polymers while exhibiting a controllable degradation rate under conventional conditions.

Interestingly, the combination of lactide and glycolide can form a new polymer known as poly(lactide-*co*-glycolide) (PLGA) which possesses great characteristics of both parent materials such as biodegradability, tunable degradation rate, good mechanical strength, excellent barrier performance, versatile processability and so on [25-27]. Guo et al. have prepared alternating PLGAs via ring-opening polymerization [28]. The controlled drug release characteristic and linear degradation indicated that these polymers can be used as an ideal carrier for mild long-acting drug delivery. Li et al. have successfully synthesized sequenced PLGAs [29]. The materials shown strong potential to use in stem cell scaffolding and drug delivery due to linear molecular weight loss profile throughout the hydrolysis.

More importantly, PLGA characteristics such as crystallinity and degradation rate can be easily controlled via copolymer composition and microstructure [30, 31]. The degree of crystallinity is decreased by about 20 % when lactide content increases from 0 to 30 %, and its orderliness will completely be collapsed once lactide content reaches 40-50 % [32]. Altay et al. synthesised PLGAs with a high glycolide content ranging from 80 to 90 % [33]. Correspondingly, degree of crystallinity increased markedly from 11 to 31 %. Rittinghaus et al. have prepared block copolymers via ring-opening polymerization in the presence of an iron guanidine complex [34]. It was not surprised when the block copolymers shown tightly ordered and highly packed structure compared to the ill-defined random series. For degradation rate, the increase of glycolidyl segment in PLGAs can speed up the initial degradation rate constants [35]. Mei et al. reported that lactide-rich PLGAs exhibited good stability in simulated body fluid, showing less than 10 % mass loss within 5 weeks [36]. In contrast, PLGAs containing 55 % lactide displayed a much faster degradation behaviour with a degradation half-life of only 4 weeks.

It is well-documented that ring-opening polymerization is an advanced technique to prepared PLGAs, especially well-defined block copolymers due to its living characteristic [37]. Since Gilding and Reed first reported the synthesis of random glycolide-rich PLGAs via ring-opening polymerization in 1979 [31], many PLGAs and their derivatives have been reported [19, 25, 38]. Although PLGA research has achieved significant progress, the development of adequate materials that possess the desired

combination of physical properties and controlled degradability for sustainable packaging applications remains many challenges [38]. Moreover, systematic studies on how microstructure and glycolide contents (50 and 90 %) influence on PLGA properties are rare. Herein, we thus prepared random and block glycolide-rich PLGAs in one-step reaction via ring-opening polymerization. Additionally, the structure-properties relationship has also been studied. The potentials of the synthesized polymers to replace traditional nondegradable plastics have been discussed as well.

2. Results and discussion

2.1. Synthesis of polymers

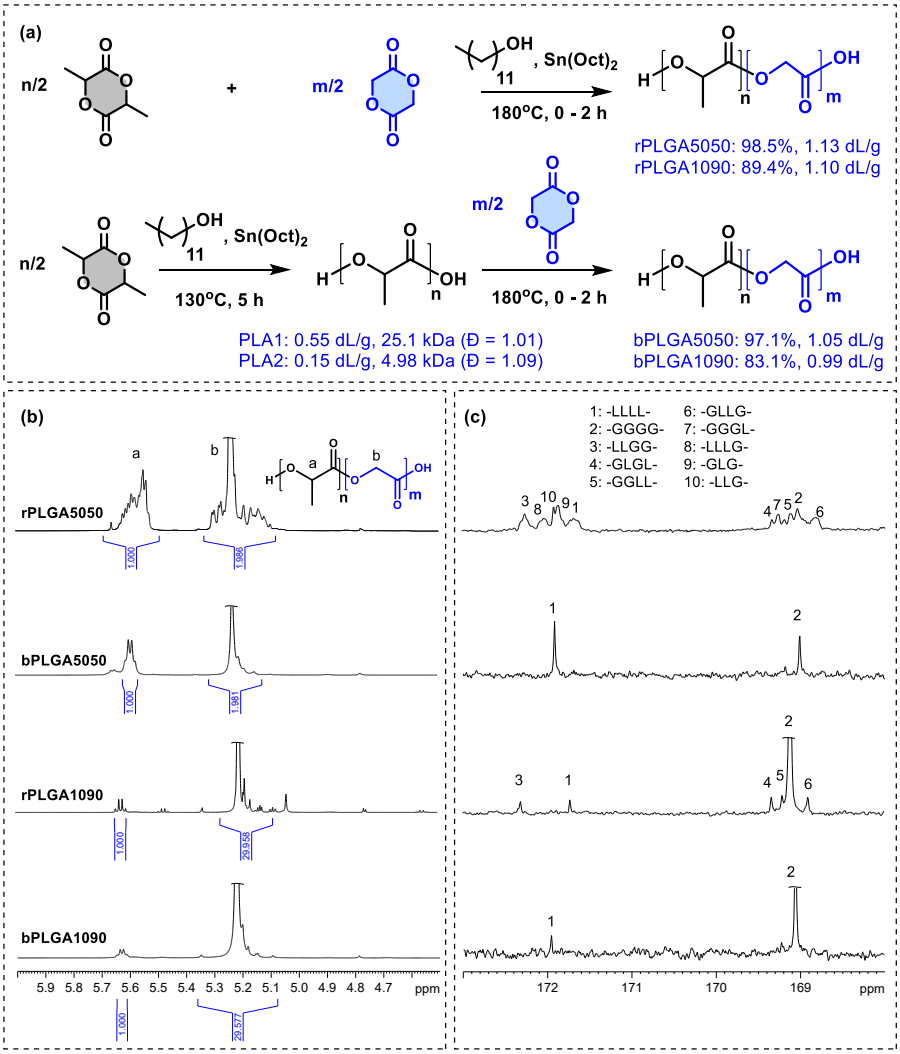


Figure 1. (a) Synthetic route, (b) ^{1}H NMR and (c) ^{13}C NMR spectra of polymers.

The synthetic route to polymers is shown in **Figure 1a**. The experiments in detail can be found in Appendices. All polymers were prepared by ring-opening polymerization of *L*-lactide and glycolide in the presence of 1-dodecanol and $Sn(Oct)_2$ as initiator and catalyst, respectively. For **rPLGA5050** and **bPLGA5050**, the feeding ratio between *L*-lactide and glycolide (P_L:P_G) was 1:1. The P_L:P_G ratio for **rPLGA1090** and **bPLGA1090** was 1:9. For the random copolymers, *L*-lactide and glycolide were copolymerized at 180 ^oC from 0-2 hours. For the block copolymers, *L*-lactide was polymerized at 130 ^oC for 5 hours to form the first block and then glycolide was added to form the second block from 0-2 hours at 180 ^oC. The crude compounds were purified by the precipitation. The solids were collected by filtration and then dried in a vacuum oven to give the polymers in a good yield ranging from 83.1-98.5 %. Intrinsic viscosity [η] of the resulted polymers are shown Fig. A.1. Polymers **rPLGA5050**, **rPLGA1090, bPLGA5050** and **bPLGA1090** shown the [η] of 1.13, 1.10, 1.05 and 0.99 dL/g, respectively. For the block copolymers, the increase of [η] after adding glycolide implied that the second block has been formed.

2.2. Polymer characterization

2.2.1. Structural characterization of polymers

The ^1H NMR spectroscopy has been used to investigate the microstructure of the polymers (**Figure 1b**). It is worth mentioning that the ^1H NMR spectra of random copolymers were significantly different compared to those of the block counterparts. The signals of random copolymers had overlapped each other. In the contrary, the peaks of block copolymers were relatively sharp indicating well-defined structure of the block polymers. From integral values of methine (δ = 5.7-5.5 ppm) and methylene (δ = 5.4-5.1 ppm) groups, the actual P_L:P_G ratio can be computed [39]. For **rPLGA5050** and **bPLGA5050**, the actual P_L:P_G ratio was equal to theoretical value of 1:1. For **rPLGA1090** and **bPLGA1090**, the calculated glycolide content was a bit lower compared to feeding ratio of 3.7 %.

The ^{13}C NMR spectra can further confirm the structure of the polymers. In **Figure 1c**, the letters *L* and *G* represent *L*-lactide and glycolide, respectively. For the block copolymers, an intense signal at about 171.9 ppm can be assigned to *L*-lactidyl segment (*LLLL*). The signal at approximately 169.1 ppm can be assigned to glycolidyl segment (*GGGG*). Importantly, the traces of heterosequences were absented in the ^{13}C NMR spectra which implied that the structure of **bPLGA5050** and **bPLGA1090** was well-organized. In case of the random polymers, the peak belonging to *L*-lactidyl segment was slightly shifted to high shielded region of about 0.2 ppm, while the signal belonging to glycolidyl segment was equivalent to that of the block copolymers. A small signal at 172.3 ppm corresponded to the link between *L*-lactidyl and glycolidyl segment (*LLGG*). Furthermore, a week signal at 169.3 ppm corresponded to the link between glycolidyl and *L*-lactidyl segment (*GGLL*). It is worth mentioning that not only homosequences, but also many heterosequences have been detected.

2.2.2. Self-assembly of polymers

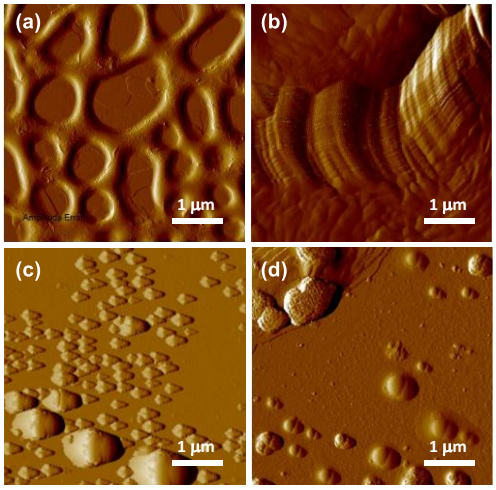


Figure 2. AFM images (a) **rPLGA5050**, (b) **bPLGA5050**, (c) **rPLGA1090** and (d) **bPLGA1090**.

AFM images of the polymers can be observed in **Figure 2**. Polymer **rPLGA5050** shown mesh-like structure. For **bPLGA5050**, it exhibited lamellar structure with *d*-spacing of few nanometres. **bPLGA1090** shown spherical structures, of which two seemed to be fused into one. The unique

morphology of the block copolymers is induced by self-assembly phenomenon. It is well-documented that the microphase separation is depended on the length of two segments [40]. In case of **rPLGA1090**, it displayed diamond-like structure with the size of about 200-400 nm. The agglomeration was observed in **rPLGA1090** as well.

2.2.3. Crystallinity of polymers

Thermal transitions of the polymers were determined by using DSC with the heating rate of 10 ºC/min (**Figure 3a,b**). Due to ill-organized structure of **rPLGA5050**, only glass transition temperature (T_g) was detected at 52 ºC. The absence of melting point (T_m) and crystallization temperature (T_c) also indicated that **rPLGA5050** was amorphous. **rPLGA1090** and **bPLGA1090** showed a T_g value of about 33 ºC. Unfortunately, the T_g values for **bPLGA5050** were not found out. **bPLGA5050** exhibited two T_m values at 144 and 213 ºC to be assigned to the melting temperature of *L*-lactidyl and glycolidyl segments, respectively. For **bPLGA1090,** two T_m values (165 and 223 ºC) were relatively greater than those of **bPLGA5050**. In case of **rPLGA1090**, only an endothermic signal at 211 ºC was recorded. In cooling cycle, **bPLGA5050, rPLGA1090** and **bPLGA1090** showed an exothermic signal at 181, 136 and 191 ºC, respectively.

Based on integral value of the endothermic peaks, enthalpy of fusion (ΔH) can be determined. The ΔH values of **bPLGA5050**, **rPLGA1090** and **bPLGA1090** were equal to 124, 118 and 183 J/g, respectively. Degree of crystallinity (χ^c) can be determined from the ΔH [25]. **bPLGA5050** gave the χ^c value of 60 %. The χ^c value of **rPLGA1090** and **bPLGA1090** was equal to 57 and 89 %, respectively. The increase of glycolide ratio leads to increasing degree of crystallinity since glycolide units are more symmetrical and less bulky structure than lactide units, allowing polymer chains to pack more regularly and form crystal more easily. It is worth mentioning that the block polymers exhibited higher degree of crystallinity random counterparts. The segmented chain architecture provides longer regular homosequences which can align and pack efficiently into order crystalline domains.

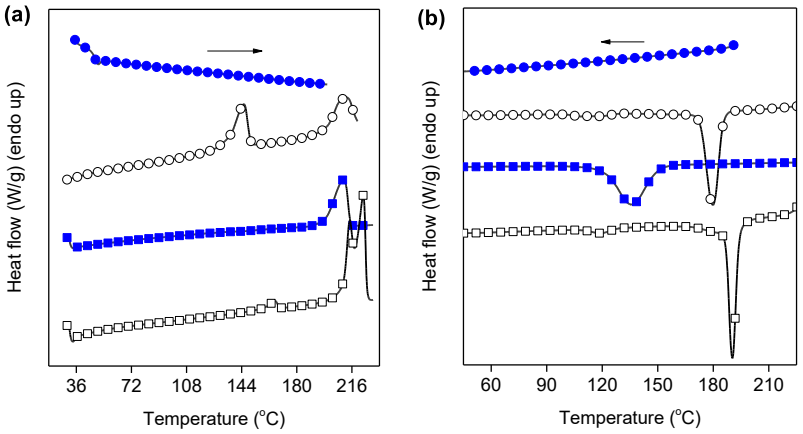


Figure 3. (a) DSC thermograms (heating cycle) and (c) DSC thermograms (cooling cycle) of **rPLGA5050** [●], **bPLGA5050** [○], **rPLGA1090** [■] and **bPLGA1090** [□].

2.2.4. Surface characteristic of polymers

The wettability of the polymers was studied by using water and diiodomethane (DIM) (**Figure 4**). The water contact angle (θ_w) of **rPLGA5050** and **bPLGA5050** was equal to 88.4 and 71.1°, respectively. The increase of glycolide ratio increased the hydrophilicity of **rPLGA1090** and **bPLGA1090** since glycolide units lacked hydrophobic methyl side groups and possessed higher polarity than lactide units. The θ_w of **rPLGA1090** and **bPLGA1090** was 64.1 and 59°, respectively. The DIM contact angle of **rPLGA5050**, **bPLGA5050, rPLGA1090** and **bPLGA1090** was 50.7, 44.3, 40.0 and 33.9°, respectively. Employing Owens-Wendt-Rabel-Kaelble model [41], the surface energy (SE) can be calculated. The SE of **rPLGA5050** and **bPLGA5050** was equal to 36.1 and 45.0 mJ/m^2, respectively. The SE for **rPLGA1090** and **bPLGA1090** was 49.9 and 54.5 mJ/m^2, respectively.

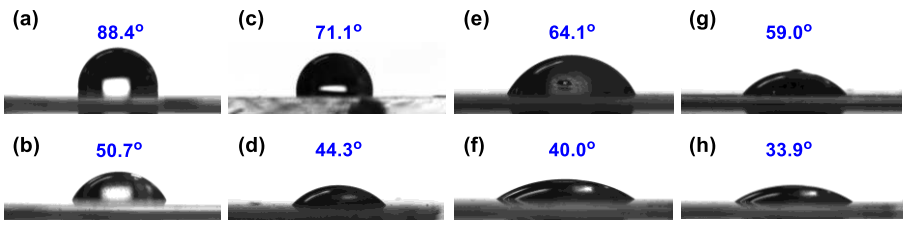


Figure 4. Water contact angle (above) and DIM contact angle (below) images of (a)(b) **rPLGA5050**, (c)(d) **bPLGA5050**, (e)(f) **rPLGA1090** and (g)(h) **bPLGA1090**.

2.3. Performances toward packaging applications

2.3.1. Thermal stability of polymers

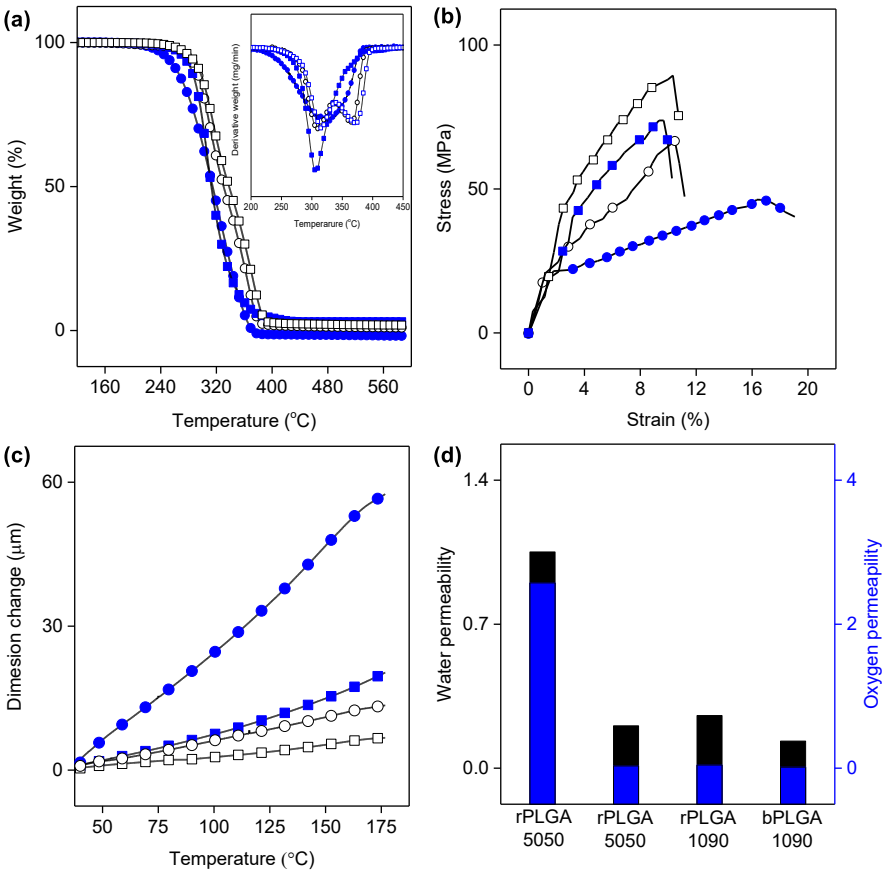


Figure 5. (a) TGA/DTG traces, (b) stress/strain curves, (c) TMA charts and (d) WP and OP of **rPLGA5050** [●], **bPLGA5050** [○], **rPLGA1090** [■] and **bPLGA1090** [□].

The thermal stability of the polymers was investigated by TGA using a heating rate of 20 ºC/min (**Figure 5a**). The DTG curves displayed that the random polymers were thermally degraded in a single-stage process while the block polymers were decomposed in a double-stage process. The first step can be assigned to the decomposition of *L*-lactidyl segment and the second one corresponded to the decomposition of glycolidyl segment. This observation can further support the well-defined structure of the block copolymers. The degradation temperature (T_d) was determined at 5% of weight loss and the results can be observed in **Figure 5a** and **Table 1**. The T_d of **rPLGA5050** and **bPLGA5050** was detected at 250 and 281 ºC, respectively. For **rPLGA1090** and **bPLGA1090**, the T_d value was recorded at 277

and 289 °C, respectively. A higher crystallinity, a better T_d can be observed since crystalline regions are ordered, densely packed and strongly bonded. These ordered domains resist chain scission and molecular motion during heating.

2.3.2. Mechanical properties of polymers

The mechanical properties of the polymers can be found in **Figure 5b** and **Table 1**. Young 's modulus (*E*) of the block copolymers was significantly higher than their random counterparts. It is worth mentioning that high glycolide content results in enhancing the *E* value. For ultimate tensile strength (UTS), **bPLGA1090** shown the highest value of 89.8 MPa. The lowest UTS belonged to **rPLGA5050** (46.7 MPa). The UTS of **bPLGA5050** and **rPLGA1090** was 66.5 and 74.7 MPa, respectively. From what has mentioned above, the mechanical properties are the function of crystallinity. The polymer chains become more closely packed and ordered, resulting in higher *E* and UTS due to stronger intermolecular interactions and improved resistance to deformation.

2.3.3. Thermomechanical stability of polymers

The TMA was used to better understand about the thermomechanical properties of the polymers (**Figure 5c** and **Table 1**). The coefficient of thermal expansion (CTE) of **rPLGA5050** was 33.1 ppm/°C. The CTE of **bPLGA5050** was much lower of 7.6 ppm/°C. The CTE of **rPLGA1090** and **bPLGA1090** was 12.1 and 3.5 ppm/°C, respectively. Crystalline regions are tightly packed and highly ordered, so that polymer chains have limited mobility during heating. Since the chains cannot move or expand freely, the material undergoes less dimensional change with temperature. In the contrary, amorphous regions contain loosely packed chains with high free volume and greater molecular motion, leading to larger thermal expansion.

2.3.4. Barrier properties of polymers

Figure 5d and **Table 1** deplete water permeability (WP) and oxygen permeability (OP) of the polymers, respectively. The WP and OP of **rPLGA5050** were 1.051 g.mm/m^2.day.bar and 2.573 g.cm^3/m^2.day.bar, respectively. **bPLGA5050** shown the better WP (0.206 g.mm/m^2.day.bar) and OP (0.206 g.mm/m^2.day.bar) compared to **rPLGA5050**. The WP of **rPLGA1090** was 0.256 g.mm/m^2.day.bar while the OP was equal to 0.044 g.cm^3/m^2.day.bar. The WP and OP of **bPLGA1090** were 0.132 g.mm/m^2.day.bar and 0.015 g.cm^3/m^2.day.bar, respectively. The transport of a gas molecule through a polymer film depends on both its solubility within polymer matrix and the ability of this molecule to diffuse through the matrix. Crystalline regions are tightly packed and highly ordered than amorphous region. Thus, they enable to prevent penetrant sorption and lower penetrant solubility.

2.4. Performance toward sustainable packaging applications

The dog bone-shaped tensile strength specimens were used in degradation experiments. The ester backbone of the aliphatic polyesters is cut via simple hydrolysis in water (**Figure 6a**) [42]. The degradability of the polymers can be found out on **Figure 7**. It is well-documented that the degradation rate depends on the morphology, molecular mass, composition ratio and crystallinity [43]. Herein, the polymers with 50 % glycolide degraded more slowly than the glycolide-rich polymers. During the first 20 days, the mass loss of the polymers containing 50 % of glycolide was very less, often known as induction period. After this period, the degradation rate was speeded up. For **rPLGA5050** and **bPLGA5050**, they were loss about 50 % of mass after 50 days. In contract, the polymers containing 90 % of glycolide were rapidly degraded without induction period. The loss mass was up to about 55 % after 20 days and rose to 70 % after 50 days. Moreover, the degradation rate of the random polymers was faster than that of the block counterparts. During degradation process, long polymer chains will be cut into shorter chains, leading to lowering viscosity (**Figure 7**).

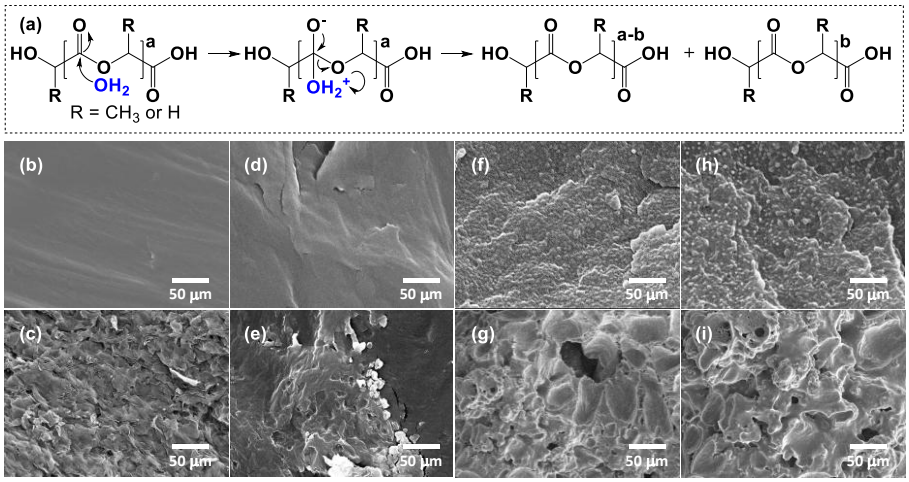


Figure 6. (a) Plausible degradation mechanism by water. SEM images of (b)(c) **rPLGA5050**, (d)(e) **bPLGA5050**, (f)(g) **rPLGA1090** and (h)(i) **bPLGA1090** before (above) and after 10 days of degradation (below).

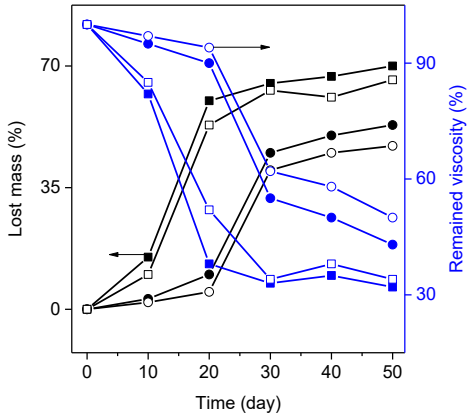


Figure 7. The plot of lost mass and remained viscosity vs day of **rPLGA5050** [●], **bPLGA5050** [○], **rPLGA1090** [■] and **bPLGA1090** [□].

As mentioned in the previous part (**Figure 4**), the glycolide-rich polymers were more hydrophilic than the polymers with 50 % glycolide. Additionally, the surface of the glycolide-rich polymers was rougher than that of the polymers with 50 % glycolide (**Figure 6,** above). Thus, water molecules can attack polymer backbone more rapidly in case of **rPLGA1090** and **bPLGA1090**. Even though **rPLGA1090** and **bPLGA1090** exhibited high crystallinity, the higher surface area can overcome the

counteracting crystallinity effect. The morphology of the specimens after exposing to water was studied by using SEM (**Figure 6**, below). The corrosion in the glycolide-rich polymers was more severe than the polymers with 50 % glycolide. The surface of **rPLGA5050** and **bPLGA5050** became rougher with some cracks observed (**Figure 6c,e**). In case of the glycolide-rich polymers, their surface was seriously corroded to form many pores (**Figure 6g,i**).

2.5. Benchmarking and implications for sustainable packaging applications

Table 1. Comparison of performances between the polymers in this work and PET.

Polymer	T_d (°C)	E (GPa)	UTS (MPa)	CTE (ppm/ °C)	WP (g.mm/ m^2.day.bar)	OP (mm.cm^3/ m^2.day.bar)	$T_{1/2}$ (day)
rPLGA5050	250	1.20	46.7	33.1	1.051	2.573	39
bPLGA5050	281	1.76	66.5	7.6	0.206	0.032	50
rPLGA1090	277	1.74	74.7	12.1	0.256	0.044	17
bPLGA1090	289	2.53	89.8	3.5	0.132	0.015	21
PET	390	1.07	45.8	70.0	0.830	4.500	73000

To evaluate the potential of the synthesized polymers as alternatives to conventional nondegradable plastics such as PET, a comprehensive comparison was carried out (**Table 1**). In this comparison, the properties of PET were collected from previous studies [11, 25, 44-46]. Although PET shows a higher T_d (390 °C), the T_d of the samples (250-289 °C) are still high enough to maintain their performance in use. The mechanical and thermomechanical properties of the samples are comparable to or even better than PET. These results suggest that the samples have sufficient rigidity and mechanical strength to withstand practical packaging and handling conditions, which is particularly important since biodegradable polymers are often considered to have weaker structural stability than petroleum-based plastics.

Furthermore, these also indicate better dimensional stability under temperature changes, reducing the risk of deformation during storage, transportation, and thermal processing. The barrier properties of the samples are greater than those of PET. Polymer films can effectively reduce the diffusion of oxygen and moisture, which are the main causes of food oxidation, quality loss, and microbial growth. Thus, the samples not only match but may even outperform PET in preserving food quality and extending shelf life. Importantly, all synthesized polymers exhibit biodegradability, confirming their ability to greatly reduce the accumulation of persistent plastic waste. Based on the above results, the synthesized polymers are highly promising sustainable alternatives to conventional nondegradable plastics, especially the block polymers.

3. Conclusions

Random and block PLGAs with different glycolide contents have successfully prepared through ring-opening polymerization. The actual P_L:P_G ratio was similar to the feeding ratio of 1:1 and 1:9. For the block series, an increase of viscosity after adding glycolide strongly proved the formation of the second segment. The ^{13}C NMR spectra were the reliable evidences to confirm well-defined structure of the block samples. Interestingly, the block materials shown a unique self-assembly characteristic while the random counterparts exhibited mesh-like or diamond-like structure. By changing ratio or structure, degree of crystallinity can be controlled, and thus polymer properties can be tailored. Except **rPLGA5050**, the others shown high degree of crystallinity from 57-89 %. The degree of crystallinity played an important role in enhancing physical properties of the polymers. Polymer with the highest crystallinity, **bPLGA1090**, shown a T_d value of 289 ºC, which was higher than the others quite far. Similarly, **bPLGA1090** possessed an UTS of up to 89.8 MPa, while this value for the other polymers ranged from 46.7-74.7 MPa. Crystal domains also acted as reinforced regions, hindering the thermal expansion of **bPLGA5050** (CTE = 7.6 ppm/ºC) and **bPLGA1090** (CTE = 3.5 ppm/ºC). They also restricted the diffusion of gas or vapour molecules through polymer matrices. For the degradability, the polymers with

50 % of glycolide degraded more difficult than glycolide-rich ones due to more hydrophobic. Additionally, the block copolymers needed longer time to fragmentize. Overall, the synthesized polymers are promising sustainable alternatives to conventional nondegradable plastics such as PET since their performances are comparable to or even better than PET.

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Conflict of Interest

The author declares no competing interests.

Author contributions

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Phan Van Hoang Khang, Do Xuan Phu, Nguyen The Duc Hanh and Tran Thi Bich Quyen. The first draft of the manuscript was written by Phan Van Hoang Khang and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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