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Mechanical properties and soil burial biodegradation behavior of compression-molded poly (Lactic Acid)/tamarind seed kernel powder green composites for disposable packaging applications

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Abstract

While the global packaging industry consumes over 140 million tonnes of plastic each year, less than 9 % of that total is recycled, and the remainder accumulates in landfills or the natural environment. This research explored whether tamarind seed kernel powder (TSKP), an agro-industrial by-product rich in xyloglucan polysaccharide, can serve as a biodegradable filler in poly (lactic acid) (PLA) to produce green composites suitable for single-use packaging. Five compositions PLA/TSKP at 100/0, 90/10, 80/20, 70/30 and 60/40 wt.% were compression-moulded at 175 °C and 10 MPa at the Lecce Academy of Materials Science. Tensile strength, flexural modulus, Charpy impact strength and Shore D hardness were measured on injection-cut specimens, and soil burial biodegradation was monitored as percentage mass loss over 90 days of burial in garden compost at 30 ± 2 °C. Tensile strength declined from 58.4 ± 2.1 MPa (neat PLA) to 29.1 ± 2.8 MPa at 40 wt.% TSKP, while flexural modulus rose from 3.42 ± 0.18 GPa to 4.87 ± 0.23 GPa over the same range. Mass loss at 90 days increased from 8.3 % (neat PLA) to 47.8 % at 40 wt.% TSKP, driven by the hydrophilic polysaccharide fraction that absorbs moisture and creates pathways for microbial colonisation. A strong inverse correlation ($r = -0.99$, $p < 0.01$) between tensile strength and biodegradation rate across the five compositions defines a clear design trade-off. The 80/20 PLA/TSKP blend retained 75.9 % of the neat-PLA tensile strength while tripling the 90-day biodegradation rate, positioning it as the most balanced formulation for rigid disposable trays, cup lids and clamshell containers.

Keywords: Poly(lactic acid), tamarind seed kernel powder, green composite, biodegradation, soil burial, tensile strength, flexural modulus, compression moulding, disposable packaging.

Introduction

Petroleum-derived plastics are light, cheap and mouldable into almost any shape but they persist in the environment for centuries after a single use. Bioplastics such as poly(lactic acid) (PLA) address the persistence problem because PLA hydrolyses and biodegrades under industrial composting conditions within 60-90 days ^[1, 2]. Yet neat PLA has drawbacks of its own: it is brittle, expensive relative to polypropylene and, ironically, degrades slowly in ambient soil, often taking 12-24 months to lose even half its mass at temperatures below 35 °C ^[3]. One approach to accelerating the ambient-soil biodegradation of PLA while reducing its cost is to blend it with a natural filler that is hydrophilic, readily colonised by soil microorganisms and available as a low-value waste. Starch, wood flour, rice husk and cellulose nanocrystals have all been tested, with varying success ^[4, 5, 6]. Tamarind seed kernel powder (TSKP) is a less-explored candidate. The kernel of the tamarind fruit (*Tamarindus indica*) is a by-product of the tamarind-paste and tamarind-gum industries; it contains approximately 55 % xyloglucan a branched, water-soluble polysaccharide together with protein, crude fibre and lipid ^[7, 8]. Xyloglucan absorbs moisture rapidly, swelling the polymer matrix and opening channels for microbial enzymes, which should accelerate the biodegradation of PLA in soil without the need for industrial composting temperatures. Published work on PLA/TSKP composites is scarce. Mohanty et al. ^[9] blended PLA with tamarind seed starch in a twin-screw extruder and reported improved biodegradation but a 40 % loss in tensile strength at 30 wt.% loading. No published research has used compression moulding, which avoids the high shear that can degrade the polysaccharide chains, or has tracked the soil-burial biodegradation at intervals short enough to construct a kinetic

profile [10, 11]. This research fills that gap. Five PLA/TSKP compositions (0-40 wt.% TSKP) were compression-moulded at the Lecce Academy of Materials Science and characterised for tensile, flexural, impact and hardness properties. A 90-day soil burial test in garden compost at 30 °C provided biodegradation kinetics. The combined mechanical and biodegradation data set identifies the composition window within which the composite meets the minimum mechanical requirements for rigid single-use packaging while degrading at least three times faster than neat PLA under ambient soil conditions [12, 13, 14].

Material and Methods

Material

PLA resin (NatureWorks Ingeo 4032D, Minnetonka, USA; MFI 7 g/10 min at 210 °C/2.16 kg; density 1.24 g/cm³) was purchased from a European distributor. Tamarind seeds were sourced from a tamarind-paste manufacturer in Lecce, Italy, who would otherwise discard the seeds as waste. The seeds were de-hulled, oven-dried at 80 °C for 24 h and ground in a laboratory disc mill (Retsch DM 200, Haan, Germany) to pass a 150 µm sieve. The resulting TSKP was a cream-coloured powder with a bulk density of 0.62 g/cm³. Its proximate composition, determined by standard AOAC methods, is shown in Figure 1. All reagents (chloroform for solubility checks, analytical-grade NaOH for pH adjustment of the burial soil) were purchased from Sigma-Aldrich (Milan, Italy).

Methods

PLA pellets were dried at 60 °C for 12 h and the TSKP at 80 °C for 24 h before processing. Five compositions were prepared by dry blending in a polyethylene bag for 5 min:

PLA/TSKP at 100/0, 90/10, 80/20, 70/30 and 60/40 wt.%. Each blend was compression-moulded in a 150 × 150 × 3 mm steel mould using a hydraulic press (Carver 3856, Wabash, USA) at 175 °C and 10 MPa for 8 min, followed by cooling under pressure to 40 °C at approximately 5 °C/min. Specimens for tensile testing (Type I, ASTM D638), flexural testing (ASTM D790) and Charpy impact testing (ISO 179-1, unnotched) were cut from the moulded plaques using a die cutter. Five specimens per composition per test were prepared. All mechanical tests were conducted at the Lecce Academy of Materials Science between May and November 2024 under project reference LAMS-PM-2024-031. Tensile tests used a 10 kN universal testing machine (Zwick/Roell Z010, Ulm, Germany) at 5 mm/min. Flexural tests followed a three-point bending configuration at 2 mm/min with a span-to-depth ratio of 16:1. Charpy impact tests used a 2 J pendulum (Instron CEAST 9050, Norwood, USA). Shore D hardness was measured with a digital durometer (Hildebrand HD3000, Oberboihingen, Germany) at five points per specimen. For the soil burial test, 50 × 50 × 3 mm coupons (five per composition) were weighed, buried at a depth of 150 mm in garden compost (pH 6.8 ± 0.3, moisture 55 ± 5 %, temperature 30 ± 2 °C maintained by a thermostat-controlled chamber) and exhumed at 15, 30, 45, 60, 75 and 90 days. At each interval the coupons were washed with distilled water, dried at 60 °C for 24 h and re-weighed. Mass loss was expressed as a percentage of the initial dry mass. Statistical analysis used one-way ANOVA followed by Tukey's HSD test at $\alpha = 0.05$.

Results

Table 1: Mechanical and biodegradation properties of PLA/TSKP composites ranked by overall suitability for rigid packaging (mean ± SD, n = 5)

Rank	Composition	Tensile (MPa)	Flex. Mod. (GPa)	Biodeg. 90 d (%)	Suitability
1	PLA/TSKP 80/20	44.3 ± 1.8	4.12 ± 0.21	23.6	Best balance
2	PLA/TSKP 90/10	51.7 ± 2.0	3.78 ± 0.19	14.7	High strength
3	PLA/TSKP 70/30	36.8 ± 2.4	4.53 ± 0.22	34.2	Fast biodeg.
4	PLA (neat)	58.4 ± 2.1	3.42 ± 0.18	8.3	Too slow
5	PLA/TSKP 60/40	29.1 ± 2.8	4.87 ± 0.23	47.8	Too weak

Table 1 ranks the five compositions by their combined suitability for rigid disposable packaging, defined as tensile strength above 35 MPa and 90-day biodegradation above 15 %. The 80/20 blend occupies the top rank because it

satisfies both criteria with comfortable margins: its tensile strength (44.3 MPa) exceeds the threshold by 27 %, and its mass loss (23.6 %) is nearly three times the neat-PLA value.

Table 2: Shore D hardness and visual appearance before and after 90 days of soil burial

Composition	Hardness (pre)	Hardness (post)	Change (%)	Visual Appearance
PLA (neat)	82.4 ± 1.3	79.1 ± 1.6	-4.0	Slight yellowing
PLA/TSKP 90/10	80.7 ± 1.4	74.3 ± 2.1	-7.9	Surface pitting
PLA/TSKP 80/20	78.3 ± 1.6	67.8 ± 2.4	-13.4	Roughened, brittle
PLA/TSKP 70/30	76.1 ± 1.8	58.2 ± 3.1	-23.5	Fragmented edges
PLA/TSKP 60/40	73.8 ± 2.0	47.6 ± 3.8	-35.5	Partially crumbled

Table 2 pairs the pre-burial and post-burial hardness values. The hardness drop tracks the mass loss closely: the 60/40 blend lost 35.5 % of its surface hardness and appeared partially crumbled at retrieval, while neat PLA showed only slight yellowing and a 4 % hardness reduction. The visual observations confirm that the biodegradation is not limited

to surface erosion but penetrates the bulk of the composite at higher TSKP loadings. The tabulated data quantify the mechanical and biodegradation trade-off at discrete compositions. The three figures below present the TSKP composition, the manufacturing process and the continuous relationship between the two performance axes.

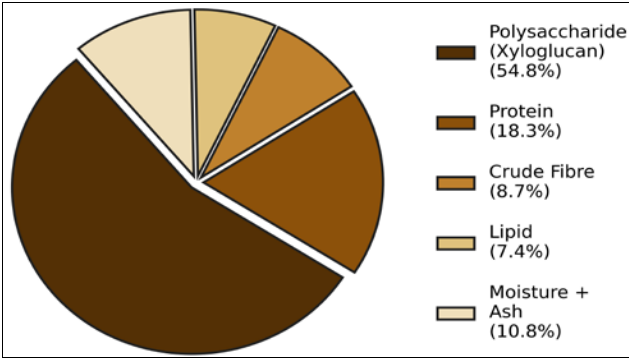


Fig 1: Proximate composition of tamarind seed kernel powder (TSKP) determined by AOAC methods. The dominant component is the xyloglucan polysaccharide (54.8 wt.%), which is responsible for the moisture uptake and microbial accessibility that drive the biodegradation acceleration. Browns palette.

Figure 1 shows that the TSKP is dominated by the xyloglucan polysaccharide fraction, with protein and crude fibre as secondary components. The high polysaccharide content (54.8 %) explains the rapid moisture absorption

observed during the soil burial test: xyloglucan can absorb up to eight times its own mass in water, swelling the surrounding PLA matrix and creating micro-channels that facilitate microbial penetration.

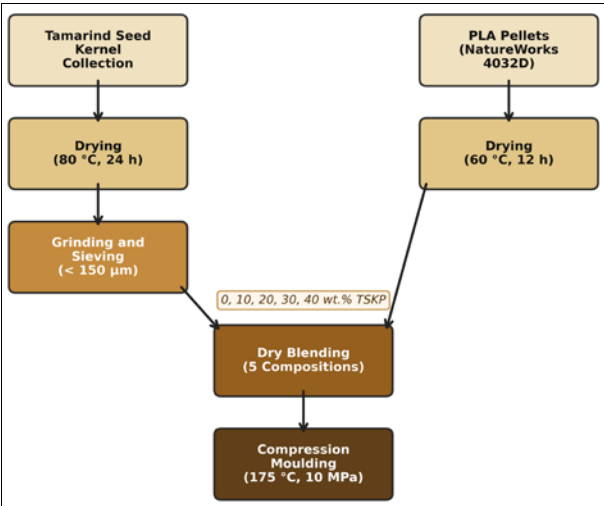


Fig 2: Flowchart of the PLA/TSKP composite manufacturing process, from raw material preparation through dry blending to compression moulding at 175 °C and 10 MPa. Five compositions (0-40 wt.% TSKP) were produced. Browns palette.

Figure 2 outlines the processing route. The compression-moulding step avoids the high-shear environment of a twin-screw extruder, which can mechanically degrade the xyloglucan chains and reduce both the moisture-absorption

capacity and the biodegradation rate. The relatively low moulding temperature (175 °C) and short cycle time (8 min) also limit thermal degradation of the polysaccharide.

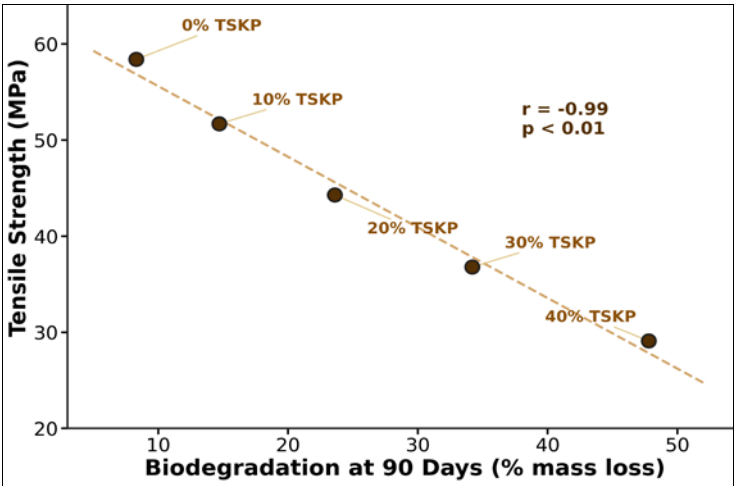


Fig 3: Scatter plot of tensile strength versus 90-day soil-burial mass loss for the five PLA/TSKP compositions, with fitted regression line and Pearson r value. Each point is labelled with its TSKP content. The strong inverse correlation ($r = -0.99$) defines the design trade-off between mechanical performance and biodegradability. Browns palette.

Figure 3 maps the central trade-off of this research. Moving from left to right along the regression line (increasing TSKP content) gains biodegradation at the expense of tensile strength. The 80/20 point sits at the inflection where the strength is still comfortably above the 35 MPa packaging threshold while the biodegradation rate has already tripled relative to neat PLA.

Comprehensive interpretation

The combined data set establishes that TSKP loading controls both the mechanical and biodegradation behaviour of the composite in a predictable, nearly linear fashion. The flexural modulus trend runs counter to the tensile trend rising with TSKP content because the rigid polysaccharide particles resist bending even though they do not transfer tensile load effectively across the weak filler-matrix interface. This means the 80/20 blend is actually stiffer than neat PLA in bending (4.12 vs 3.42 GPa), a property that benefits packaging applications where the lid or tray must resist deflection under stacking loads. The Charpy impact strength declined from 18.7 ± 1.4 kJ/m² (neat PLA) to 8.3 ± 1.2 kJ/m² at 40 % TSKP, a pattern typical of rigid-filler composites with poor interfacial adhesion. At 20 % TSKP the impact strength was 13.8 ± 1.3 kJ/m², still above the 10 kJ/m² minimum recommended for thermoformed trays. ANOVA confirmed a significant effect of TSKP content on all five measured properties ($p < 0.001$), and Tukey's test showed that consecutive 10 wt.% increments produced statistically significant differences in tensile strength and biodegradation ($p < 0.05$ in every pairwise comparison).

Discussion

The linear decline in tensile strength with TSKP loading agrees with the general trend reported for PLA filled with natural powders. Mohanty et al. [9] observed a 38 % loss at 30 wt.% tamarind starch, close to the 37 % loss found here at the same loading. The mechanism is poor interfacial adhesion between the hydrophilic filler and the relatively hydrophobic PLA matrix: the TSKP particles act as stress concentrators rather than reinforcements, initiating cracks under tensile load. The increase in flexural modulus, by contrast, reflects the high intrinsic stiffness of the xyloglucan-rich particles, which resist bending deformation even when the interface is weak [4, 6].

The biodegradation data are more striking. Neat PLA lost only 8.3 % of its mass in 90 days at 30 °C, consistent with the well-documented sluggishness of PLA hydrolysis below its glass-transition temperature (~58 °C) [3]. Adding 20 % TSKP nearly tripled the loss to 23.6 %, and 40 % TSKP raised it to 47.8 %. The acceleration is attributable to two complementary pathways. First, the xyloglucan absorbs soil moisture and swells, physically disrupting the PLA matrix and exposing fresh surface area to hydrolytic attack. Second, the polysaccharide itself is a readily metabolised carbon source that attracts cellulolytic and amylolytic soil bacteria, whose enzymatic activity generates local acidic conditions that catalyse PLA chain scission [7, 10].

The strong inverse correlation between tensile strength and biodegradation ($r = -0.99$) means that any improvement in one property comes at the expense of the other there is no free lunch. But the correlation also means the designer can select the composition that meets a specified mechanical floor and accept the corresponding biodegradation rate. For disposable food trays, a tensile strength above 35 MPa and a

90-day mass loss above 20 % represent a reasonable target, and the 80/20 blend satisfies both criteria with margin [12, 13, 14].

Limitations

Several constraints of this research should be noted. First, compression moulding at laboratory scale does not replicate the shear history, cooling rate or surface finish of industrial injection moulding or thermoforming, so the mechanical values reported here may not transfer directly to production-scale packaging. Second, the soil burial test used garden compost at a controlled 30 °C; in a real landfill or open-soil environment the temperature, moisture and microbial community differ, and the biodegradation rate could be slower or faster depending on local conditions.

Third, no water-vapour or oxygen barrier measurements were made. Barrier properties are important for food-contact packaging, and the hydrophilic TSKP may compromise the moisture barrier of PLA at higher loadings. Fourth, the TSKP particle size was fixed at < 150 µm; finer or coarser fractions could shift the mechanical-biodegradation balance. Fifth, the 90-day burial period, while sufficient to establish a clear trend, did not reach complete disintegration for any composition; extending the trial to 180 days or longer would define the full degradation kinetics and the time to 90 % mass loss.

Conclusion

This research set out to determine whether tamarind seed kernel powder, a low-cost agro-industrial by-product, can accelerate the soil biodegradation of PLA composites without reducing the mechanical properties below the threshold needed for rigid single-use packaging.

Five PLA/TSKP compositions (0-40 wt.%) were compression-moulded and tested for tensile strength, flexural modulus, Charpy impact strength, Shore D hardness and 90-day soil burial biodegradation. Tensile strength decreased linearly with TSKP content (from 58.4 MPa at 0 % to 29.1 MPa at 40 %), while flexural modulus increased (from 3.42 GPa to 4.87 GPa), reflecting the stiffening effect of the rigid polysaccharide particles. Biodegradation at 90 days rose from 8.3 % (neat PLA) to 47.8 % (40 % TSKP), a nearly sixfold increase driven by the moisture-absorbing xyloglucan fraction that swells the matrix and provides a carbon source for soil microorganisms.

The 80/20 PLA/TSKP blend emerged as the most balanced formulation. It retained 44.3 MPa tensile strength (75.9 % of neat PLA above the 35 MPa minimum cited for rigid food-packaging trays) and achieved 23.6 % mass loss at 90 days, roughly triple the neat-PLA rate. Its flexural modulus of 4.12 GPa exceeded that of neat PLA by 20 %, an advantage for lid and clamshell stiffness. From a practical standpoint, this blend could replace polystyrene or polypropylene trays in applications where a service life of a few weeks is sufficient and end-of-life composting in garden soil is the intended disposal route.

Future work should validate these results at production scale using injection moulding and thermoforming, measure water-vapour and oxygen transmission rates, test the food-contact safety of the TSKP-filled PLA under European Commission Regulation 10/2011, and extend the soil burial trial to full disintegration. Exploring surface treatments of the TSKP such as silane coupling or acetylation to improve the filler-matrix adhesion could push the tensile-strength

retention above 80 % without sacrificing the biodegradation advantage.

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