

Contents lists available at ScienceDirect

Materials Today Communications





"Film-stacking method as an alternative Agave tequilana fibre/PLA composite fabrication"

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ARTICLE INFO

Keywords: Post-harvesting waste Natural fibres Bio-materials Film-stacking Mechanical properties

ABSTRACT

This paper presents the fabrication of an Agave tequilana Fibre (ATF) bio-composite by film stacking method and an experimental investigation on preliminary mechanical properties; namely tensile, flexural, impact, and water uptake. Randomly oriented bio-composite samples were made from both, untreated and surface treated ATF collected from a tequila distillery waste stream. The ATF were surface treated using NaOH, and Pectate lyase to improve fibre properties and adhesion. The samples were prepared using three fibre loadings, 20, 40 and 60 wt%. Randomly oriented ATF mats were pre-impregnated with poly (lactic acid) (PLA) before bio-composite stacking and press moulding. Untreated fibre samples were processed and kept as a reference. The morphology of the fracture surfaces was analysed through optical and environmental scanning electron microscopy (ESEM). The quasi-static tensile tests revealed that the modulus of the bio-based composite materials is up to \sim 10% lower than that of the neat PLA. Flexural strength values observed at 60 wt% of fibre content were within the ranges of 76.10–77.0 MPa for treated samples with a flexural modulus of 3.36–3.76 GPa, whereas impact strength presented a reduction of \sim 32–35%. Generally, all measured properties presented reduction in strength with the increase of fibre loading.

1. Introduction

The increased environmental awareness and sharply risen prices of fossil fuels have encouraged the use of renewable carbon-based materials. Bio-based materials have helped decrease fossil fuel dependency and the related environmental impacts towards a reduced carbon footprint economy [1–3]. However, some challenges such as processing, mechanical properties, cost and recycling are yet to be addressed [4].

Bio-based materials made from renewable agricultural and forestry feedstocks have witnessed considerable attention as a possible solution to the ever-growing environmental concerns and as an answer to fossil fuel dependency [5–7]. Biopolymers such as PLA have experienced a renaissance in recent years with new technological developments and their wider introduction into mainstream use [8–10]. Higher market penetration of bio-based plastics has placed PLA as one of the first players with an approximate 3 Mt capacity [11]. The scenario is promising, it shows a fast-growing demand for natural fibres and bio-based materials due to rising demand for more sustainable products

and the increasing penetration of bio-based materials in the industry [12].

New biopolymers have faced major challenges to get into the market because prices are still higher than their oil-based counterparts, for which supply chain and production has been gradually optimised since they appeared more than 60 years ago [13]. Adding natural fibres to biopolymers can potentially lower manufacturing materials cost; and in some cases it has been shown to increase biopolymers performance too [14].

The use of agro-industrial by-product materials (i.e., natural fibres) from natural and annually renewable sources is mainly driven by their eco-friendliness and due to economic reasons [15,16]. Natural fibres already play a fundamental role in the "green" and circular economy [17] based on their energy efficiency, use of renewable feedstocks, low CO_2 manufacturing processes, weight reduction, recyclability and waste minimisation [18,19].

Natural fibre-based composites have several advantages over synthetic counterparts in targeted applications not requiring high load-

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https://doi.org/10.1016/j.mtcomm.2022.103853

Received 28 March 2022; Received in revised form 16 May 2022; Accepted 13 June 2022 Available online 16 June 2022

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bearing capabilities [1]. Composites based on surface-modified natural fibres have in general better mechanical and physical properties [20, 21]. The potential applications of natural fibre-based composites in housing, automotive [22,23], packaging [24] and other low-cost, high volume applications is enormous [25,26]. Bio-based composites with surface-modified natural fibres, in general, display better mechanical and physical properties due to improved adhesion and enhancement of polar interactions at the fibre/matrix interface [27–29]. Hence, a complete matrix fusion to facilitate fibre impregnation is a vital requirement [30,31]. Additionally, new knowledge on ATF/PLA-based composites will interease the use of agro-industrial by-products as raw material, and intensify research on its use within the rising green and circular economy.

Agro-industrial waste materials can be reused and upcycled for the development of new and greener composites [32–34]. Lignocellulosic fibres obtained as a by-product from the Tequila industry are a good example of these materials and a potential feedstock source for new bio-based composite developments [35,36]. Tequila by-products amount up to 300,000 t, concentrated in a small region in Mexico [37]. Despite upcycling of these by-products has proven to be feasible [38,39]; residual agave leaves are still used as soil nourishment, and the bagasse is either treated as compost or disposed of by incineration or landfill [40].

Initial evidence from a bio-based material from the agro-industrial by-product fibres from tequila production has been reported using extrusion moulding as a compounding method [41]. However, fibre content % (w/v) and relevant mechanical properties reported are low due to the compounding method used. In consequence, the development of a higher fibre content biocomposite material with General Purpose Polystyrene (GPPS) comparable properties has yet to be reported.

Film stacking method represents an advantageous alternative to extrusion and injection moulding when high fibre volume is required without the loss of fibre properties because of the high shear strain that reduces the fibre aspect ratio during the compounding process [42]. Film stacking method consists of heating and compressing a stack of alternated layers of polymeric matrix and fibre mats during a determined amount of time as shown in Fig. 1.

In this study, a preliminary investigation on the fabrication of randomly oriented ATF/PLA based composites is presented. Combining film stacking and compression moulding, bio-based composite samples were prepared using pre-impregnated ATF mats and PLA film as a matrix as it is described in the following sections.



2. Materials and methods

Tequila by-product ATF were supplied by "La Fortaleza" distillery plant in Jalisco, Mexico. Materials used included analytical grade NaOH from Acros Organics; Pectate lyase enzyme (Scourzyme®) from Novozymes, pelletized PLA (Ingeo[™] Biopolymer 2003D) by Natureworks[™], and dichloromethane from Proton Scientific. All characterisation techniques and methods used for preparing the samples throughout the work are described below.

2.1. Fibre treatments

ATF with a mean length of 10 mm were pre-washed with distilled water to remove loosely physisorbed compounds from tequila production and oven-dried at 60 °C for 24 hrs. Dry fibres were further exposed by immersion to alkali and enzymatic treatments. Alkali treated fibres were immersed 180 min in 8% (v/w) NaOH solution, while enzyme treated samples were exposed to 0.4% (v/w) of pectate lyase enzyme in relation to fibre content during 180 min. Details from every treatment have been reported elsewhere [36]. Untreated ATF samples were kept as control. Different sets of treated and untreated fibres were rinsed with distilled water and oven-dried after treatment at 60 °C for 24 hrs. and kept in sealed desiccators to control relative humidity (RH) due to the hygroscopic nature of the fibres. Fibres moisture content after drying was below ~1.5 wt%. Different sets of specimens will be referred hereafter as alkali treated fibres (AKF), enzyme treated fibres (ENF) and untreated fibres (UNF).

2.2. Biocomposite manufacturing

ATF/PLA biocomposite manufacturing was carried out using a 4 step process as summarised in Fig. 2. Randomly oriented ATF mats were prepared during the first step of the process by spreading 10 mm length (mean) ATF onto a vibration plate until an even distribution was achieved. During a second stage, mats were pre-impregnated with a 15% (w/w) PLA/dichloromethane solution and dried at 23 °C at 50% of RH. Laying up was done by stacking by hand the pre-impregnated ATF mats and PLA films according to 20%, 40%, and 60% (w/w) of fibre content onto a metallic frame. In the final stage, the manufacturing of the randomly oriented ATF/PLA biocomposite specimens was carried out by compression moulding on a 40 T thermal press at 160 °C and at a pressure of 55 MPa. Residence time was calculated to keep the melt flowing and to prevent degradation during all the runs, for at least 10 min. After pressing, the frame was quenched under room conditions. Samples are denoted hereafter as AKF, ENF and UNF for alkali, enzyme and untreated specimens with 20%, 40% and 60% (w/w) of fibre content respectively. Neat PLA plates were also press moulded under the same conditions to keep as control.

Specimens for mechanical testing were cut out from composite and PLA plates using a water jet cutting machine and according to specific ASTM standard D638–10, D790 and D4812. Samples were then ovendried at 60 °C for 24 hrs. and kept in sealed desiccators before mechanical testing.

2.3. ESEM and optical microscopy

ATF/PLA biocomposite specimens were examined pre- and postfailure using an optical microscope Nikon Eclipse E600 at 5x and 10x magnification, a macro at x0.75 and x1.5, and an Environmental Scanning Electron Microscope (ESEM) (FEI XL30) to analyse the test results and investigate possible correlations between processing conditions, mechanical performances and type of fracture.

2.4. Mechanical testing

Quasi-static mechanical properties were evaluated from tensile,



Fig. 2. Schematic from ATF/PLA composite manufacturing using pre-impregnation process.

impact and three-point bending tests. Tensile testing was carried out on type 1 specimens according to ASTM D638–10 standard on a calibrated Instron 5500 R EM with a 100 kN load cell and a crosshead speed of 2 mm/min at room temperature (21 $^{\circ}$ C) [43]. Ultimate Tensile Strength (UTS) was calculated using:

$$\sigma = \frac{L}{A} \tag{1}$$

where σ is the stress, *L* is the maximum load, and *A* is the specimen crosssectional area. Strain was also calculated using the following equation:

$$\varepsilon = \frac{\Delta L}{L_0} = \frac{L - L_0}{L_0} \tag{2}$$

where ΔL is the change in gauge length, L_0 is the initial gauge length, and L is the final length. Gauge length was directly measured with an extensioneter. Young's Modulus (*E*) was calculated by dividing the stress (σ), by the strain (ε), in the elastic portion of the stress-strain curve.

Three-point bending flexural test was performed according to ASTM D790 standard using a 5 kN load cell and a crosshead speed of 1 mm/ min [44]. Flexural stress (σ) was calculated according to:

$$\sigma = \frac{3FL}{2bd^2} \tag{3}$$

where *F* is the maximum load on the load deflection curve, *L* is the support span, and *b* and *d* correspond to the width and thickness from the specimen. Flexural modulus (E_f) was calculated using:

$$E_f = -\frac{L^3m}{4bd^3} \tag{4}$$

where *m* is the gradient of the initial straight-line portion of the loaddeflection curve. Impact testing was executed according to ASTM D4812 standard for unnotched Charpy impact on a Zwick pendulum impact test machine with a 1 J hammer. Impact strength was calculated using the following equation:

$$I = \frac{E_c}{hb} \times 10^3 \tag{5}$$

where E_c is the corrected energy absorbed by the specimen; h and b are the thickness and width of the specimen respectively. Flexural and impact tests were performed on a flatwise direction. Before testing, specimens (see Fig. 3) were oven-dried at 60 °C for 24 hrs. and kept all the time in sealed desiccators due to the hygroscopic nature from the phases. 14 specimens were tested for every material grade.

2.5. Water absorption

Water Absorption at Saturation (WAS) was determined for ATF/PLA



Fig. 3. 60% (w/w) AKF/PLA biocomposite specimens. Top: specimen used for tensile test. Middle: specimen used for three-point flexural bend test. Bottom: specimen used for impact test.

biocomposite materials. WAS is expressed as the increase in weight percent of the material according to:

$$MC = \frac{w_1 - w_0}{w_0} \times 100$$
(6)

Where (*MC*) is the moisture content, (w_0) is the mass of dry sample and (w_1) is the mass after exposure.

Specimens were first dried in an oven for 24 hrs at 60 $^{\circ}$ C and then placed in a desiccator to stabilise at room temperature (23 $^{\circ}$ C). Upon cooling, samples were weighed in an analytical balance to the nearest 0.001 g and immerse in sealed beakers containing distilled water at room temperature. The weight difference was measured at several intervals of time by removing samples from the beaker, wiping them dry and weighting them to the nearest 0.001 g until equilibrium. Three replicates for every set of samples were assessed and MC calculated.

3. Results and discussion

3.1. Biocomposite processing

3.1.1. Density

14 biocomposites samples were manufactured with a fibre wt% ranged from 20%, 40% and 60% As it was expected from the additions of a less dense ATF, density of composites decreased gradually with the increasing of ATF content. In general, a maximum density reduction of \sim 28% was presented at fibre contents of 60%. ATF/PLA biocomposite density values are within the ranges of GPPS as depicted in Fig. 4. However, ATF/PLA bio-based material at 60 wt% of fibre content indicated a density lower than 1 g/cm3. These ranges are lower than GPPS and PLA. Thus, the use of ATF/PLA bio-based material could represent energy savings when transport is involved.

The impregnation process formed consistent and uniform ATF/PLA



Fig. 4. ATF/PLA material density mean values. Error bars are smaller than the markers in some cases. Standard error < 0.02 in all cases.

mats (Fig. 5a), losing uniformity only near the borders of the mat (Fig. 5b). During the manufacturing process ATF and PLA were subjected to a pressure-temperature cycle in only one occasion whereas at compounding by extrusion moulding the materials were exposed twice at shear and temperatures cycles affecting negatively the thermomechanical properties of the compound. However, residence time required to ensure a uniform compounding was considerably longer than other conventional methods such as extrusion [41]. The main constraint during processing was controlling the hygroscopic nature of ATF and PLA due to moisture release during the process. Despite fibres and matrix were subjected to a moisture removal stage and kept in sealed desiccators during the time prior to press moulding, some traces of moisture release in the form of minuscule bubbles were present (Fig. 6b).

The pre-impregnated mats, temperature sensitivity of the fibres (well above processing temperature) and PLA, along with the residence time during processing allowed an almost uniform flow and fibre-matrix interaction as it can be seen in Fig. 6a. Consequently, an appropriate fibre/matrix interphase can be expected through a film stacking-press moulding method with pre-impregnated ATF mats. However, the improved interphase interaction is given as a result of mechanical interlocking between treated fibres and matrix [36].

3.1.2. Biocomposite structure

Microscopy observations revealed the scale variations in diameter and morphology of AKF; some effects of fibre moisture and processing as discussed in the previous section are also presented (Fig. 6b). Although there is an apparent uniform flow of matrix around the fibre, and void formation was minimal, some air trapped bubbles were present in small amounts and only opposite to the press moulding direction. These imperfections can be one of the main factors accounted for the reduction in mechanical properties as crack propagation may be prone to start in those areas. Notwithstanding this was observed in small areas and close to the mould edges, it was finally attributed to the mould design not allowing sufficient matrix and air flow across the mould section. Therefore, a better mould design, an improved control of PLA viscosity within ATF thermal degradation threshold might reduce this effect.

3.2. Mechanical testing

125 tensile tests were conducted using specimens of different "weight percentage" and treatment. Fig. 7 shows the behaviour of the mean stress-strain curve for UNF specimens in comparison with neat PLA. As observed, specimens with the highest fibre wt% presented a reduced elongation before failure. ATF/PLA composites that contained 60 wt% of alkali treated ATF have shown superiority in tensile and bending properties by ~13% and ~150% respectively in comparison to untreated specimens.

Fibre loading generally degraded the mechanical properties of the ATF biocomposite material as reported in other studies where PLA was used as matrix [41,45,46]. Although in some cases an improvement was observed, in particular for samples that were manufactured with 40 wt% and 60 wt% of treated fibre content. In these cases, it appears that the removal of non-cellulosic components from ATF specimens contributed positively to fibre/matrix interfacial interactions leading to a better matrix to fibre stress transfer in comparison with the UNF specimens. However, the reinforcing effects decreased in line with the addition of higher ATF wt% content due to the brittle and by-product nature of ATF. ATF/PLA composites presented a matrix's inherent brittle failure without knecking during tensile test (Fig. 7).

Despite the improved behaviour presented by ATF/PLA biocomposite materials using treated ATF at 40 wt% and 60 wt% content, the reduced stress transfer from the matrix to the ATF is thought to be result of an incomplete interfacial interaction promoted by the inherent flaws on the by-product natural fibres and a non-perfect adhesion between ATF and PLA. However, adhesion and enhancement of polar interactions at the fibre/matrix interface can be improved [27–29,47] through the use of compatibilizers [48].

This reduction in mechanical properties has been shown before by ATF/PLA biocomposites compounded by extrusion moulding [41]. The decrease in properties was attributed to the low fibre aspect ratio resulted from the inherent crushing of fibres by the extrusion moulding



Fig. 5. a) Section from an ATF mat impregnated with PLA/dichloromethane solution. b) Impregnated ATF/PLA Mat dimensions.



Fig. 6. a) Section from a 60% w/w ATF/PLA composite at 2x magnification showing randomly oriented ATF. b) sample section indicating the formation of air bubbles.



Fig. 7. Mean stress-strain curves of 20%, 40% and 60% (w/w) UNF/PLA biocomposites. Standard error < 0.91 in all cases.

process as referred previously by Keller (2003) [49], coupled with the initial damage fibres have suffered during tequila production.

15 specimens of each group of samples were tested for three-point bending flexural test. Fig. 9 present the flexural modulus results as a function of the fibre volume content and surface treatment applied. Flexural and tensile modulus of the ATF/PLA biocomposite at the maximum fibre wt% content remained approximately the same, without any detriment to original matrix modulus as shown in Figs. 8b and 9. The maximum tensile and flexural stress of ATF/PLA biocomposites containing treated fibres were better than the UNF only at 40 and 60 wt %, with improvements of ~13% and ~150% respectively as presented



Fig. 9. Fibre content effect on Flexural modulus for ATF/PLA biocomposites in comparison with GPPS.

in Figs. 10 and 11. Generally, neat PLA shows better tensile and flexural strength than its composites, these findings are in agreement with works previously reported [46,50].

Impact strength values of ATF/PLA biocomposite materials are presented in Fig. 12. 15 tests for each set of specimens were conducted to investigate the effect of surface treatments and fibre content on the impact strength of biocomposite materials. Although treated ATF increased the amount of energy required to pull-out and fail fibres in comparison to UNF, the impact strength has shown reductions as an effect of the increased ATF content. An improvement of ~25% for AKT samples at 60 wt% fibre content in comparison to untreated samples was also observed. However, fracture analysis presented predominant fibre pulled-out over fibre failure without big traces of PLA matrix remaining on the fibre surface. In general, these results show that the ATF/PLA material properties are improved by mechanical interlocking between



Fig. 8. Tensile properties for ATF/PLA biocomposites manufacture by film stacking method. a) Fibre content effect on UTS for ATF/PLA composites. b) Fibre content effect on Young's modulus for ATF/PLA biocomposites.



Fig. 10. Effect of treatments on Ultimate Tensile Strength, Young's modulus and Strain on ATF/PLA biocomposites manufactured by film stacking method. Standard error bars are smaller than the markers in some cases.



Fig. 11. Effect of surface treatment and fibre content on flexural strength and flexural modulus for ATF/PLA biocomposites (flatwise) in comparison with general purpose polystyrene. Standard error bars are smaller than the markers in some cases.



Fig. 12. Impact strength of ATF/PLA biocomposites in comparison with GPPS and PLA.

fibre and matrix when treated ATF are used. Although this improvement is not significant, further treatments, compatibilizers and better processing conditions controls could be considered to improve the toughness of the ATF/PLA biocomposite materials.

The most important factor originating the decreasing trend in mechanical properties of the ATF/PLA materials lies in the flaws presented at the interface of the material and the structure and by-product character of the ATF. After the apparently improved interfacial interaction due to pre-impregnation process and surface treatment, the ATF fibre structure still remains the weakest link in the system. However, ATF/PLA biocomposite materials at higher fibre ratio contents yet have comparable properties to GPPS.

3.3. Water absorption

Water uptake of ATF/PLA biocomposites was found to increase with fibre loading, and more evidently after 20 wt% of ATF content. This increase was attributed to the physical and chemical changes occurred at ATF during treatments and strictly correlated to the degree of delignification and removal of non-cellulosic materials. However, AKF/PLA composites at 60 wt% content presented the major difference with a reduction in water uptake of ~15% in comparison with untreated samples as shown in Fig. 13. In this case the difference in water uptake is thought to be result of an improved matrix-fibre interaction that reduced the hydrophilic nature of alkali treated fibres [51,52].

ESEM and Optical microscopy images obtained at the fracture surfaces of tensile test specimens shown in Figs. 14 and 15, presented pulled-out fibres. However, it can be seen that the surfaces of the pulledout fibres are not totally clean. These observations suggest a low adhesion between the fibres and the matrix, as presented in other natural fibre/PLA based composites [46]. Nevertheless, treated samples depicted an improvement in adhesion contrary to the reduced interfacial interaction presented by untreated samples as can be corroborated by Bax and Müssig J [53].

In the case of untreated samples (Fig. 14b), pull-outs occurred more frequently than with alkali treated fibres. AKF/PLA biocomposites have shown better fibre-matrix bonding due to the removal of hydrophobic materials (i. e., lignin, pectin and extractives) as shown in Fig. 14a. This better compatibility provided better mechanical properties than untreated samples due to a more evident interlocking between fibre and matrix. However, PLA melting viscosity and matrix flow within the mould appeared to be insufficient to facilitate the proper impregnation of AKF.

Observations performed on mechanical-tested ATF/PLA biocomposites highlighted that treated samples showed a reduction in fibre pull-out with better matrix impregnation as compared to the UNF samples. Fracture processes of ATF/PLA materials presented yield crazing in accordance to Jiang et al. [54]. Fracture surface of biocomposite materials was brittle due to the less plastic deformation properties inherent to the PLA matrix. This brittle fracture was accompanied by longitudinal fibre pull-out found for the majority of fibres arranged in a transverse direction to the crack plane (Fig. 14); furthermore, these fibres were not able to fully contribute to the crack resistance of the material.



Fig. 13. Water absorption properties of ATF/PLA biocomposites. Standard error bars are smaller than the markers in some cases.



Fig. 14. ESEM micrographs of tensile tested biocomposites. a) 60% w/w AKF/PLA. b) 60% w/w UNF/PLA.



Fig. 15. Fracture section from a 60% w/w ATF/PLA biocomposites showing delamination after failure at the tensile test.

4. Conclusions

This paper has reviewed the mechanical properties of an ATF/PLA based material manufactured though film staking method and the associated constraints encountered in bringing together by-product natural ATF and PLA to develop a bio-based material for non-structural applications. Also, the effect of different fibre ratios and surface treatments on the material mechanical properties has been investigated.

The pre-impregnated mats and compression moulding process used for this research extended ATF capability of absorbing PLA, hence, improving interfacial interaction and allowing higher ATF ratio content in comparison with results previously presented where compounding via extrusion moulding was used [41]. Despite, these open mould processes produce composites with high levels of voids (porosity), PLA impregnation and flow can be improved by a better control of PLA melting viscosity within the narrow processability window offered by ATF and through mould design optimisation. The compounding method used in this work provided improved mechanical properties in comparison to compounding by extrusion, due to the incorporation of longer fibres contrary to the low fibre aspect ratios present in extrusion moulding process. Furthermore, higher fibre wt% content achieved through this method offers the possibility of a material with acceptable properties using as much as possible a raw material source with a marginal cost that was previously considered waste.

There was an inherent compatibility between ATF and PLA; and despites surface modification helped to some extent to overcome intrinsic problems in the use of ATF by showing better mechanical properties in comparison with untreated ATF, results are almost negligible when economic and environmental benefits are sought. From an environmental point of view, the improvements obtained through surface modification treatments might be considered little convenient, encouraging its use only for specific applications. Other ways of promoting interfacial interaction between ATF and PLA (e. g., melt viscosity control) to achieve a composite material with better mechanical properties suitable for more demanding applications might prove more economical, environmental, and efficient.

Results obtained in this work give an overview about the possibilities of an ATF/PLA bio-based material by comparing its properties against GPPS. In particular, the best UTS values were reported for 20% (w/w) UNF/PLA composites with 43.47 MPa followed by 40% (w/w) AKF/PLA with 34.76 MPa and 60% (w/w) AKF/PLA with 32.54 MPa respectively. All biocomposites have shown the same decrease trend in properties with the increase of ATF content regardless the surface treatment used. However, and considering the high % content of discarded material used, characteristics and benefits are comparable or within the range to those of GPPS and other PLA based composites where natural fibres are specifically produced for composite applications ⁴⁶. Furthermore, the

water uptake of treated ATF/PLA biocomposites was found to be reduced for treated samples, being \sim 15% of water uptake reduction the major difference observed for AKF/PLA biocomposites at 60 wt% content in comparison with untreated samples.

Data has shown that ATF/PLA biocomposites can be considered as materials for non-load bearing applications such as packaging, containers, crates and other targeted applications where high strength is not sought, but environmental benefits for a fast-moving consumer product are the main concern. Due to ATF characteristics and by-product origin, there are still many stages through to harvest-procurement and processing at which variability in properties (degradation) can be introduced. Therefore, a significant opportunity window for improvement is presented.

CRediT authorship contribution statement

Omar Huerta-Cardoso: Conceptualization, Investigation, Writing – original draft. **Isidro Durazo-Cardenas:** Validation, Formal analysis. **Phil Longhurst:** Visualization, Supervision. **Adriana Encinas-Oropesa:** Methodology, Writing – review & editing, Supervision, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This research was funded by the Mexican Government through the Consejo Nacional de Ciencia y Tecnologia (CONACyT) and the Secretaria de Educacion Publica (SEP). Distillery "La Fortaleza" is gratefully acknowledged for the support in providing the material and access to their facilities.

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