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FULL PAPER

Coloration Technology

Effect of organic biocolourants on lightfastness and mechanical properties of polylactic acid

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Abstract

Most modern colourants are acquired from petroleum-based sources or mining, which often causes environmental degradation. Consequently, alternative colouring solutions are sought, such as using biocolourants, which are organic colourants from bio-based sources. Although biocolourant research is prevalent in the textile dyeing field, biocoloration of thermoplastics remains relatively unexplored. Thus, our study presents lightfastness and mechanical properties of polylactic acid (PLA) biocoloured with biochar, kraft lignin, indigo from *Isatis tinctoria*, madder extract from *Rubia tinctorum*, and weld extract from *Reseda luteola*. Indigo, madder extract, and weld extract are common bio-based textile dyes, whereas biochar and lignin are more known as easily accessible filler additives than as colourants. Stemming from promising results with bio-based indigo in our previous studies, this study broadens the scope with new colourants and analysis methods. In this study, mixtures of biocolourant powder and PLA granules were heated to a viscous state, blended inside an injection screw, and injection moulded into plastic specimens. The PLA-biocolourant specimens were inspected with scanning electron microscopy (SEM) and subjected to ASTM D638 tensile test. Furthermore, the specimens were exposed to ISO 105-B02 lightfastness test under xenon arc light. Before and after xenon light exposure, the specimens were analysed with visual inspection, reflectance spectrophotometry, hyperspectral imaging, and Fourier-transform infrared (FTIR) spectroscopy. The results showed that most of the biocolourants produced an even colour with adequate lightfastness. Moreover, lignin, biochar, and madder extract substantially improved photodegradation resistance and tensile properties of PLA. Thus, further development of biocolourants, especially lignin, as functional PLA additives is recommended.

1 | INTRODUCTION

Organic colourants derived from natural sources, hereafter referred as biocolourants, are traditional alternatives to modern petroleum-based synthetic colourants.[1,2] Due to renewable origin of biocolourants, published research generally describes them as more environmentally friendly alternatives to synthetic colourants.[3–5] Due to recent surge of scepticism towards such general statements, the undeniable eco-potential of natural colourants

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is no more taken as granted but has inspired many academics to focus on biocolourant sustainability in practical use.[4,6–8] Consequently, several recent studies have reported that in many cases, biocolourants can indeed be more sustainable than their synthetic counterparts, especially if the raw materials are obtained from industrial side streams.[8,9] Residual streams from agriculture can also be sustainable sources for anthocyanin, quinone, and carotenoid type biocolourants.[10] Finally, even though high land use requirements prevent mass production of most plant-cultivated biocolourants, Pattanaik et al demonstrated in 2020 that indigo dye production from *Indigofera tinctoria* can be feasible life cycle-wise compared to synthetic indigo production.[11]

Despite the sustainability potential of biocolourants, their worldly consumption is currently less than 1% of the ever-growing consumption of synthetic colourants.[12] Several reasons contribute to the popularity of synthetic colourants, for instance, they are substantially cheaper to produce in large volumes compared to biocolourants.[13] Moreover, the synthetic colourants offer a complete colour palette with good fastness properties and even colourant quality for almost every dyeing application.[14] In contrast, natural colourants provide a considerably narrower colour palette with limitations in colour intensity and dyeing capabilities altogether.[14–16] Because of these limitations, the use and production of biocolourants is mostly done in relatively small scale by either hobbyist dyers or particularly creative entrepreneurs.

Due to the increasing sustainability demands in the twenty-first century, many researchers have made determined efforts to work their way around the limitations and non-durable reputation of biocolourants.[15,17] A common approach has been to introduce novel colourants extracted from locally abundant raw materials, thus highlighting the availability and scale-up potential of the biocolourant.[18–20] Another approach has been to enhance either colour or fastness properties of known biocolourants, thus making them better suited for specific applications or more durable in general.[21–23] Furthermore, in the textile dyeing field, many researchers have also experimented with bio-based mordants to achieve broader colour palettes and stronger biocolourant–fibre interactions.[24–26] Additionally, a ubiquitous but important research approach in textile field is to thoroughly test fastness properties of selected biocolourants, proving that they can fulfil industrial demands.[27,28] Finally, the trendiest approach is to experiment with functional biocolourants, which are designed to provide added value to the coloured product beyond just the colour itself.[29,30] For example, the colourant can improve microbial resistance or ultraviolet (UV) protection of textiles or generate antioxidative properties for food products.[31–33] The antioxidative and UV-resistant properties are especially prominent in polyphenolic biocolourants, such as flavones, anthraquinones or polylignols.[34–36]

In the continuous quest for more sustainable, durable, and affordable biocolourants, most of the research focuses on textile dyes, food colourants, or cosmetics. However, the increased knowledge about biocolourants has greatly expanded the scope of their potential applications. Consequently, many studies in more non-traditional fields of biocolourant research have been conducted. For instance, plant-based indigo has been used as a colourant for linseed oil-based paints and varnishes, resulting in a wood coating with substantial UV protection.[37] Furthermore, fair lightfastness properties for coloured paper were achieved with various plant-based dyes in 2018.[38] Regarding the polymer field, Velho et al encapsulated natural carmine, turmeric, indigo and annatto dyes inside a silica gel matrix to improve lightfastness of the dyed polyvinyl chloride (PVC). However, the dye encapsulation had no substantial effect to lightfastness of the coloured plastics.[39]

Plastics are among the most colourant-consuming materials used in everyday consumer products and packaging. Naturally, plastics cannot sidestep the sustainability requirements of the modern materials. Thus, bio-based plastics like polylactic acid (PLA) have gained interest as materials for more sustainable plastic products. Colouring bio-based plastics with bio-based colourants, however, is still a relatively new topic with limited amount of publications. In 2019, Fajraoui et al coloured PLA with a dye extracted from *Acacia cyanophylla* plant, achieving a material with good lightfastness properties.[3] Furthermore, biocoloured PLA packaging films have been made either by mixing the colourant inside PLA matrix via extrusion or by attaching the colourant on top of the film with polymeric hosts.[40,41] PLA fibres have also been dyed with natural colours, for instance, Baykuş et al studied modification of PLA fibres to enhance their dyeability with alizarin, lawsone and indigo.[42]

Probably due to relatively low number of publications about plastic biocoloration, fastness or mechanical properties of biocoloured materials with likely upscalable production has been given little consideration.[43] To shed some light on the practical side of plastic biocoloration, this study presents findings on lightfastness and mechanical properties of PLA coloured with bio-based colourants. Five biocolourants were chosen based on their distinctive chemical characteristics: indigoid-type indigo, anthraquinone-type madder extract, flavone-type weld extract, lignan-type kraft lignin, and pigment charcoal. The indigo, madder extract and weld extract are largely produced plant-based dyestuffs originating from *Isatis tinctoria*, *Rubia tinctorum* and *Reseda luteola*, respectively.[5]

Biochar and kraft lignin are less familiar as biocolourants, but more known as abundant forest industry side streams. Both biochar and lignin are poorly soluble to water or ethanol, which encourages their use as pigments rather than dyes, whereas the other colourants in this study are mostly used as dyes with water or ethanol base. Nevertheless, high availability and low market value of lignin and biochar could make them appealing to use as pigments despite their somewhat limited applicability and colour palette. PLA, in turn, is a bio-based thermoplastic, which is commonly used for instance in three-dimensional (3D)-printing and food packaging applications.[44]

2 | MATERIALS AND METHODS

2.1 | Materials

Transparent PLA granules of injection moulding grade 3052D (molecular weight (M_w) = 116,000 g/mol, melting temperature (T_m) = 145–160°C) were obtained from NatureWorks LLC. Dried water–ethanol extract of madder with alizarin as main constituent was purchased from Greening Ltd. Similarly, dried water–ethanol extract of weld consisting of luteolin with traces of apigenin and chrysoeriol was purchased from Greening Ltd. Purified kraft lignin was obtained from UPM-Kymmene Ltd. Natural indigo pigment was obtained from Natural Indigo Finland Ltd. and pretreated by soaking in excess hydrochloric acid (2 mol/L) and thoroughly rinsing with distilled water afterwards. Additionally, biochar was produced at Häme University of Applied Sciences by pyrolysing a locally harvested wood chip mixture, which contained approximately 50% of alder chips and 50% of birch chips. The pyrolysis process was conducted in 600°C for 20 min, after which the obtained biochar was ground and dried overnight in 55°C, resulting to coarse biochar powder with < 10% moisture content.

2.2 | Methods

Each of the five colourants were dried in 50°C for 24 h and ground to powder for 10 min with IKA A 10 impact mill. After grinding, the colourant powders were thoroughly mixed with pre-dried PLA granules in the weight ratio of 1:1000 to achieve distinctive but not oversaturated colour with each colourant. The colourant–PLA mixtures were injection moulded into two different shapes of sample rods: dogbone-shaped specimens suited for ASTM D638 tensile testing and smaller, rectangular specimens for other analysis methods. In addition to the coloured mixtures, sample rods were moulded from uncoloured PLA in the same manner. Conditions of the injection moulding are presented in Table 1. Notably, the nozzle temperature was chosen to be high enough to achieve good mouldability but low enough to avoid potential colourant degradation.

TABLE 1 Conditions for injection moulding.

Moulding equipment	Arburg Allrounder 370 A
Screw size	30 mm
Injection speed	45 mm/s
Injection pressure	1500 bar
Feed zone 1 temperature	135°C
Barrel zone 2, 3, 4 temperatures	145, 155, 165°C
Nozzle temperature	175°C
Mould temperature	30°C

After injection moulding, tensile tests for the dogbone-shaped specimens were performed according to standard ASTM D638 using a ZwickRoell tensile tester. The test was performed to eight parallel specimens of each plastic to determine their average tensile strength, Young's modulus, and elongation at break with a 95% confidence interval. Visual properties of the smaller samples were analysed by visual observation and scanning electron microscopy (SEM). Furthermore, ImageJ image processing software was used to approximate median particle diameter of the colourants based on high-resolution images acquired from optical microscope using 8× magnification. Approximately 1000 visible colourant particles were counted from each image and included in the particle size calculation.

Spectral properties of the specimens were measured with Datacolor 600 reflectance spectrophotometer. To achieve a tangible colour expression, the spectral data used to determine various CIE colour coordinates: lightness parameter L^*, green-red colour parameter a^*, and blue-yellow colour parameter b^*. Additionally, CIE Y reflectance parameter of each specimen was measured with the reflectance spectrophotometer over both white and black background. To indicate colour strength of each specimen, contrast ratio (CR) was calculated from the CIE Y values measured over the white and black background according to Equation (1).

$$\mathrm{CR} = \frac{Y_b}{Y_w} \times 100\%. \quad (1)$$

To extend from visual wavelength range, hyperspectral reflectance of each specimen was assessed with Specim IQ portable hyperspectral imaging system. Furthermore, to access chemical properties of the specimens, Fourier-

transform infrared (FTIR) spectroscopy with attenuated total reflection (ATR) was measured with Shimadzu IRSpirit. Post-analysis, the specimens were subjected to ISO 105-B02:2016 lightfastness test in a Heraeus Suntest CPS+ Xenon arc light chamber. After the lightfastness test, the samples were analysed again by visual observation, reflectance spectroscopy, hyperspectral imaging, and FTIR-ATR to detect possible changes caused by the light exposure.

3 | RESULTS AND DISCUSSION

3.1 | Injection moulding

Injection moulding PLA–colourant mixtures into plastic rods produced samples with consistent dimensions and smooth surface. SEM inspection of the injection moulded plastics showed no evidence of abnormal surface defects or deformations. Furthermore, median diameter of 3–8 μm for colourant particles was observed in all coloured plastics. SEM images of all plastics are presented in Figure 1.

Regarding colour and visual appearance, the plastics coloured with lignin, madder extract, and weld extract had smooth and even colour with partially translucent characteristics and no visible colourant particles. On the contrary, plastics coloured with biochar or indigo appeared opaquer but contained visually distinguishable colourant particles. In biochar-coloured plastic, the colourant particles were evenly distributed and barely distinguishable by the human eye, presenting a deep, mostly uniform, and highly opaque black colour. However, the blue indigo-coloured plastic contained substantial clusters of colourant powder inside the plastic, presumably due to agglomeration of the colourant particles during the injection moulding process. Despite colourant agglomeration, the indigo-coloured plastic exhibited deeper colour compared to the samples coloured with lignin, madder extract, or weld extract. Photographs of the injection moulded sample rods are shown in Figure 1.

3.2 | Lightfastness

Indoor lightfastness performance of the coloured plastics was determined by xenon light exposure through a window glass filter according to modified ISO 105-B02 standard. Figure 1 presents appearance of each plastic before and after 168-h light exposure period and Figure 2D shows lightfastness ratings of the plastics. As seen in Figure 2D, uncoloured and biochar-coloured plastics had the best lightfastness performance by visual evaluation, closely followed by the lignin-coloured plastic. Surprisingly, only the indigo-coloured plastic showed poor lightfastness performance to a human observer, even though indigo is generally known as a highly light-resistant colourant. To reinforce the visual lightfastness evaluation, Table 2 presents the total colour differences calculated from CIELab colorimetric parameters according to Sharma et al.[45] Similar to the visual evaluation, no substantial colour difference was detected in the biochar-coloured or uncoloured plastics after the lightfastness test, whereas the indigo-coloured plastic underwent a

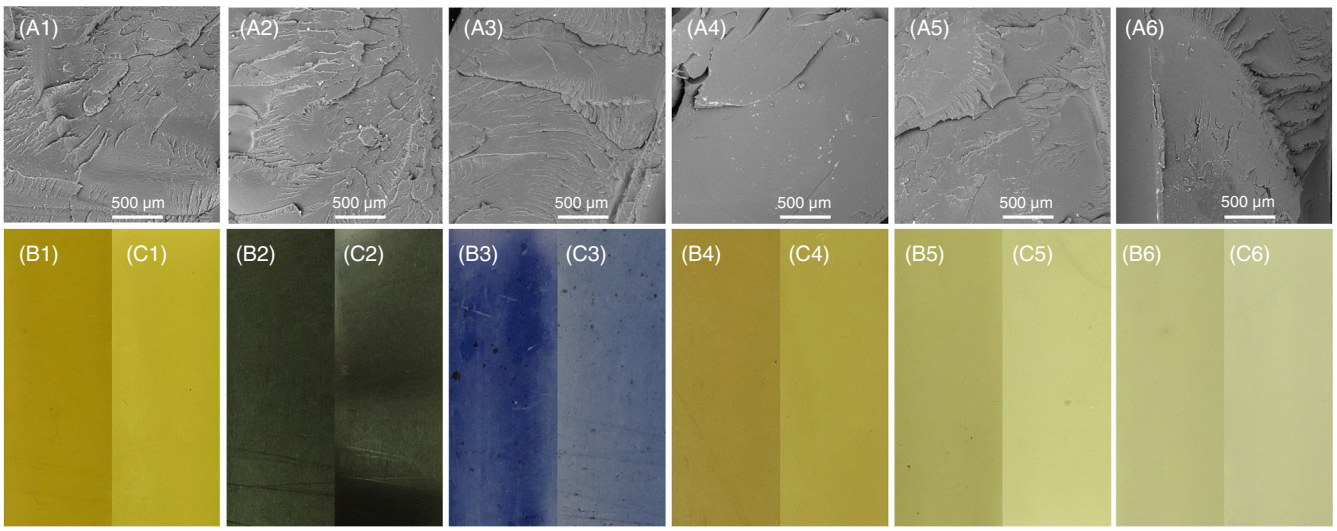


FIGURE 1 Images of the injection moulded polylactic acid rods. Top row: (A) SEM images with 100× magnification. Bottom row: Photographs before (B) and after 168 h (C) of xenon light exposure. Colourants: 1, madder extract; 2, biochar; 3, indigo; 4, lignin; 5, weld extract; 6, no colourant.

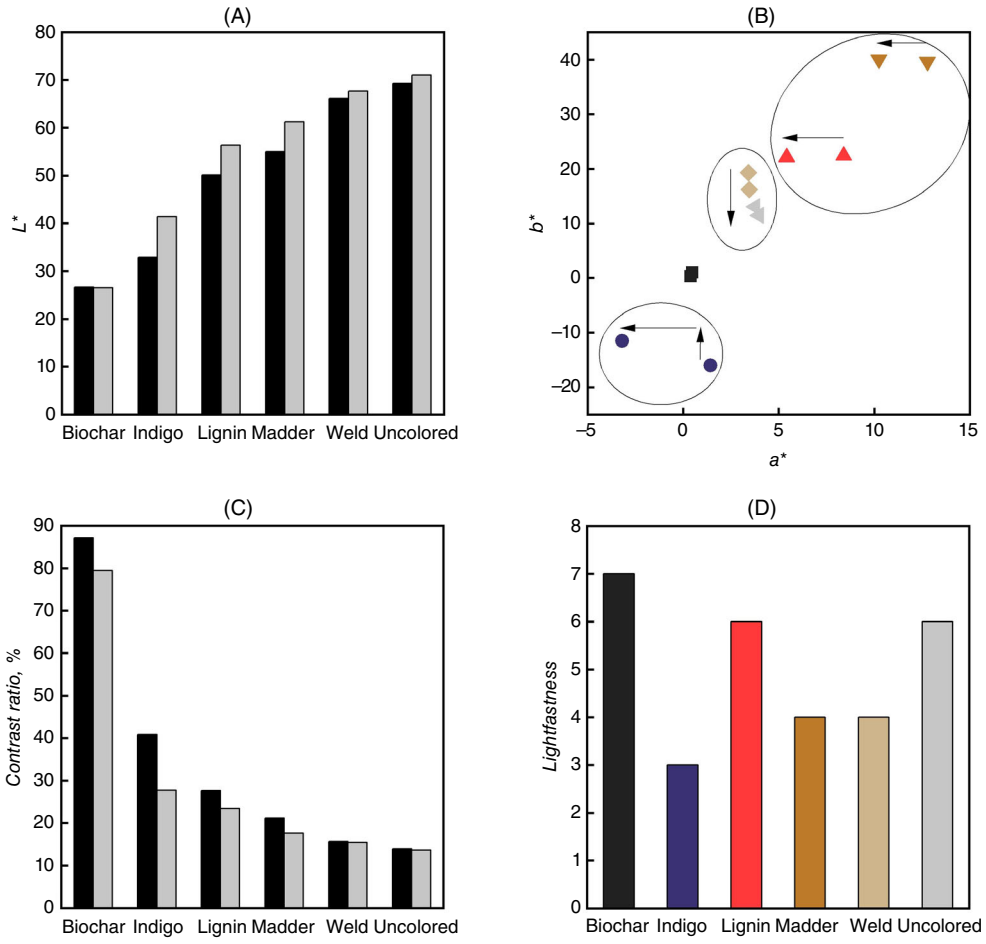


FIGURE 2 Colorimetric properties of the plastics before and after xenon light exposure. (A) CIE lightness L^*, (B) CIE hue parameters a^* (green-red) and b^* (blue-yellow), (C) contrast ratio and (D) lightfastness value. Colours in (A) and (C): Black = unexposed specimen and grey = exposed specimen. Colours in (B) and (D): Black = biochar, blue = indigo, red = lignin, bronze = madder, ecru = weld, and grey = uncoloured specimen.

TABLE 2 Total colour changes for each plastic after 168 h of xenon light exposure.

Colourant	Total colour difference (CIEDE2000)
Biochar	0.7
None	1.7
Weld extract	2.2
Madder extract	6.0
Lignin	6.5
Indigo	9.6

major colour change. However, compared to the lightfastness results from visual evaluation, the lignin-coloured plastic showed a more pronounced colour difference, which was comparable to the change in madder-coloured plastic. In contrast, the colorimetric evaluation showed only an insignificant colour difference for the weld-coloured plastic despite achieving only moderate lightfastness rating in visual assessment.

Based on both visual and colorimetric evaluation, PLA coloured with the relatively inert biochar pigment had higher lightfastness compared to the plastics coloured with polyphenolic madder, weld, and lignin. The difference in lightfastness is explained by the tendency of polyphenolic colourants to absorb UV light and enter an excited high-energy state.[46] As the excited colourant molecules return to the ground state, the released energy may break intramolecular bonds, leading to colourant degradation.[47,48] On a more positive note, the absorptive behaviour of polyphenol colourant may reduce the photoenergy load towards the plastic matrix and consequently inhibit its photodegradation.[49] Importantly, photodegradation rate of a polyphenol colourant is not related to its photoprotective ability by default. For instance, some polyphenols, such as lignin, can controllably release the absorbed photoenergy and mostly retain their original structures while offering UV-protection for the coloured material.[50]

The individual CIELab colorimetric parameters and contrast ratios for each sample before and after light exposure are presented in Figure 2A–C. Figure 2A presents the CIE lightness values before and after the light exposure. During the light exposure period, the indigo-coloured plastic exhibited the highest increase in CIE lightness followed by madder- and lignin-coloured samples. The light exposure did not substantially affect CIE lightness of any other plastics.

In many cases, continuous light exposure may decrease colour intensity and change hue of the exposed specimen. Hue and intensity of a colour in specific lighting can be presented with its CIE green-red colour parameter a^* and CIE blue-yellow colour parameter b^*. Figure 2B presents the colour parameters a^* and b^* in a coordinate system to illustrate effects of light exposure to hue and colour intensity of the coloured plastics. Figure 2B shows that the light exposure decreased a^* value of the madder- and lignin-coloured samples, shifting their yellow-red colour towards pure yellow and decreasing their overall colour intensity. In contrast, a drop in the b^* value was the main effect observed with yellow weld-coloured and uncoloured samples, decreasing their colour intensity, and minimally shifting their colour towards redder hues. The largest changes in hue and colour intensity were again observed in the indigo-coloured plastic, which experienced a substantial loss in colour intensity and shifted its colour from blue-red to blue-green hues. Finally, the light exposure had no substantial effects to either hue or colour intensity of the biochar-coloured plastic.

The CR values in Figure 2C illustrate opacity of the plastics before and after the light exposure period. Figure 2C shows that biochar produced substantially opaquer colour than the other colourants in this study, even though all coloured plastics contained exactly 0.1 w% of colourant. Conversely, weld produced the most translucent colour, which was only slightly opaquer than the original colour of uncoloured PLA. The CR values also show that opacity of all samples decreased during the light exposure period. Unsurprisingly, the indigo-coloured plastic had the highest loss in opacity among all coloured samples. The weld-coloured plastic was the only coloured specimen with insignificant opacity loss, most likely due to highly translucent initial colour of the unexposed plastic.

3.3 | Spectral properties

Reflectance spectra of the samples in the 360–700 nm wavelength region are presented in Figure 3A. Figure 3A shows that light exposure led to increased reflectance for all plastics except the biochar-coloured one, for which no changes in reflectance was observed. Furthermore, all plastics except the biochar-coloured one presented a wide reflectance peak starting from the yellow 560–590 nm wavelength region and growing steadily towards the red 620–700 nm wavelength region. During light exposure, this peak was slightly shifted towards shorter wavelengths, which explains the red-to-yellow hue changes presented earlier in Figure 2. Despite the peak shift, relative proportions of any reflectance peaks did not substantially change during the exposure period.

Reflectance spectra of the plastics in hyperspectral wavelength range of 800–1000 nm were measured to detect any visually unnoticeable degradation caused by light exposure. Figure 3B shows that light exposure slightly decreased hyperspectral reflectance of all coloured plastics except the biochar-coloured one. However, any significant changes for peak locations or shapes were not observed. Notably, plastics with high initial reflectance in the hyperspectral region suffered a proportionally higher reflectance loss across the region compared to the plastics with low initial reflectance. Moreover, substantial signal noise was observed for individual pixels near the maximum edge of spectral measuring range at 1000 nm, which may affect the final result. Thus, based on hyperspectral reflectance alone, none of the studied colourants seemed to conclusively affect polymer degradation during light exposure.

3.4 | Chemical degradation

To extend from the reflectance examinations, FTIR-ATR spectroscopy was used to investigate possible chemical degradation of the plastics during light exposure. The FTIR-ATR spectra of the plastics in before and after light exposure are presented in Figure 4A. The abundant –C–O ester bonds of PLA are presented by vibrating stretch peaks at 1081, 1130, and 1181 cm^{-1}.[51] Furthermore, $–CH_3$ bending groups can be seen at 1453 cm^{-1} and –CH– deformation groups at 1362 and 1382 cm^{-1}.[52] Also, a substantial presence of –C=O carbonyl groups is indicated by a strong stretching peak at 1747 cm^{-1} and a small bend peak at 1267 cm^{-1}.[53] Finally, a small peak at 957 cm^{-1} combined with absence of a peak at 921 cm^{-1} indicates mostly an amorphous structure of PLA.[54–56]

As shown in Figure 4A, the light exposure provoked substantial growth of the –C=O stretch peak at 1747 cm^{-1}. Multiple studies have previously shown that increased levels of –OH and –C=O groups in PLA after light exposure indicate photodegradation by Norrish Type II reaction.[57–60] This photon-initiated reaction breaks the ester bonds of PLA, forming two separate polylactides with COOH and –OH end groups.[51,61,62] Other plausible photodegradation mechanism proposed by Janorkar et al leads to formation of diketone and carboxylic acid end groups via formation of hydroperoxide derivative.[63,64] However, contrary to multiple previous studies regarding both aforementioned reaction mechanisms, emergence of neither anhydride band at 1845 cm^{-1} nor carboxylic acid bands at 1720 or 3290 cm^{-1} was not observed.

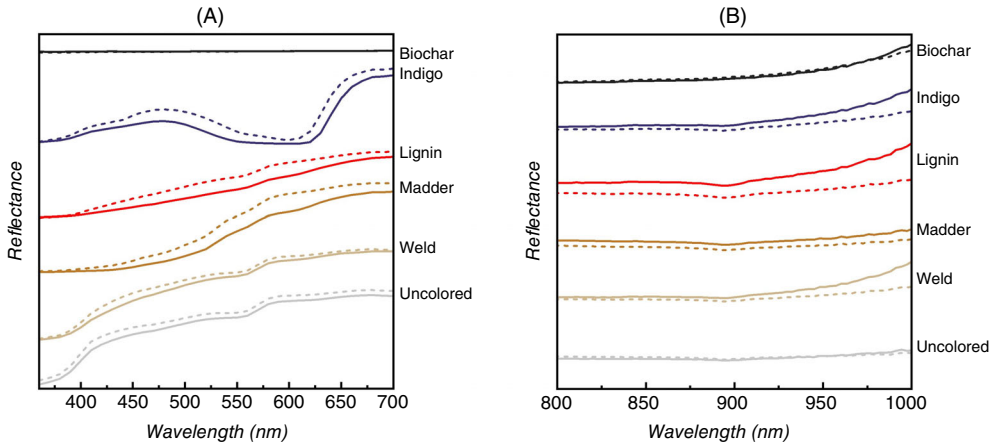


FIGURE 3 Reflectance of the plastics in (A) visible 360–700 nm wavelength region and (B) hyperspectral 800–1000 nm wavelength region. Solid line = unexposed sample and dashed line = exposed sample. Colours in the figure: Black = biochar, blue = indigo, red = lignin, bronze = madder, ecru = weld, and grey = uncoloured polylactic acid.

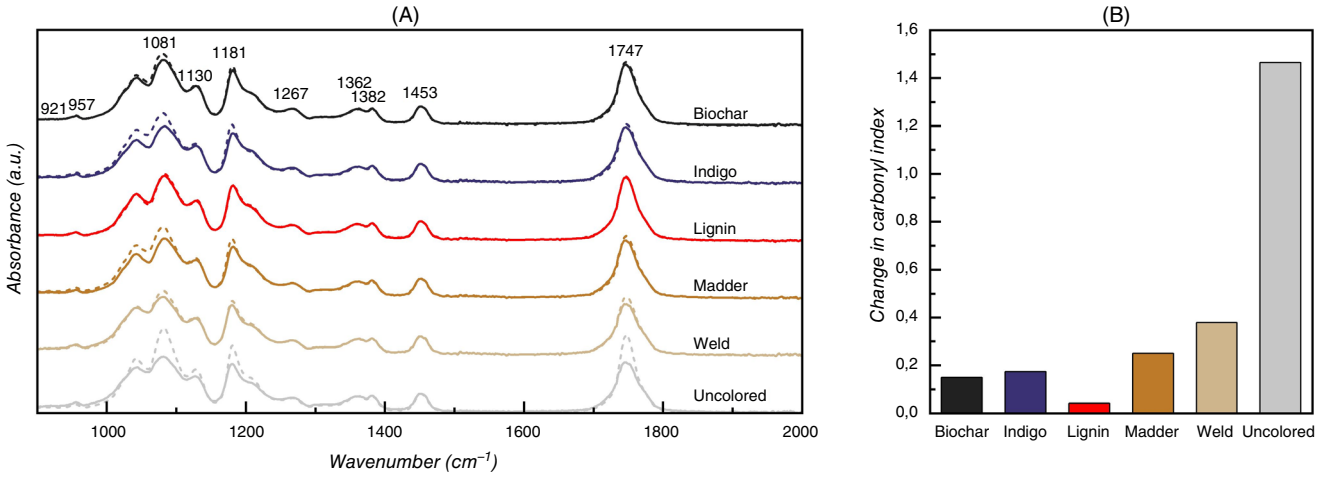


FIGURE 4 (A) Fourier-transform infrared-attenuated total reflection (FTIR_ATR) spectra of the plastics before (solid line) and after (dashed line) xenon light exposure. (B) Light-induced changes in carbonyl indexes of the plastics. Colours in the figure: Black = biochar, blue = indigo, red = lignin, bronze = madder, ecru = weld, and grey = uncoloured polylactic acid.

Due to the absence of emerging absorption bands, degradation of the polymer was quantified by determining its carbonyl index, which indicates relative amount of carbonyl groups inside the polymer. For PLA, the carbonyl index can be presented as relative intensity of the photosensitive –C=O stretch peak at 1747 cm^{-1} to the stable $-CH_3$ peak at 1453 cm^{-1}.[57–59,65] Multiple studies have shown that an increase in carbonyl index corresponds to photo-oxidative degradation of PLA.[58–60] Thus, the light-induced changes in carbonyl index (ΔC_i) for all plastics was calculated from the absorbance peak intensities before and after light exposure with the following equation:

$$\Delta C_i = \frac{A(C=O)}{A(-CH_3)} - \frac{A_0(C=O)}{A_0(-CH_3)}. \quad (2)$$

The ΔC_i values caused by light exposure are shown in Figure 4B. As expected, uncoloured plastic degraded the most due to lack of protective colourant. On the contrary, lignin-coloured plastic was almost fully resistant to photo-oxidative degradation. The substantial protection from lignin is most probably due to a large number of UV-absorptive functional groups in lignin macromolecules.[34,66–68] Besides lignin, all other colourants offered substantial degradation resistance as well.

3.5 | Tensile properties

All plastics were subjected to tensile test to determine whether colourant addition had any effect on mechanical properties of the plastic. According to the average ultimate tensile strengths presented in Figure 5A, the highest tensile strength was achieved with the lignin-coloured plastic, followed by madder and biochar. Moreover, lignin, madder and biochar substantially strengthened the uncoloured PLA, which suggests that besides colouring

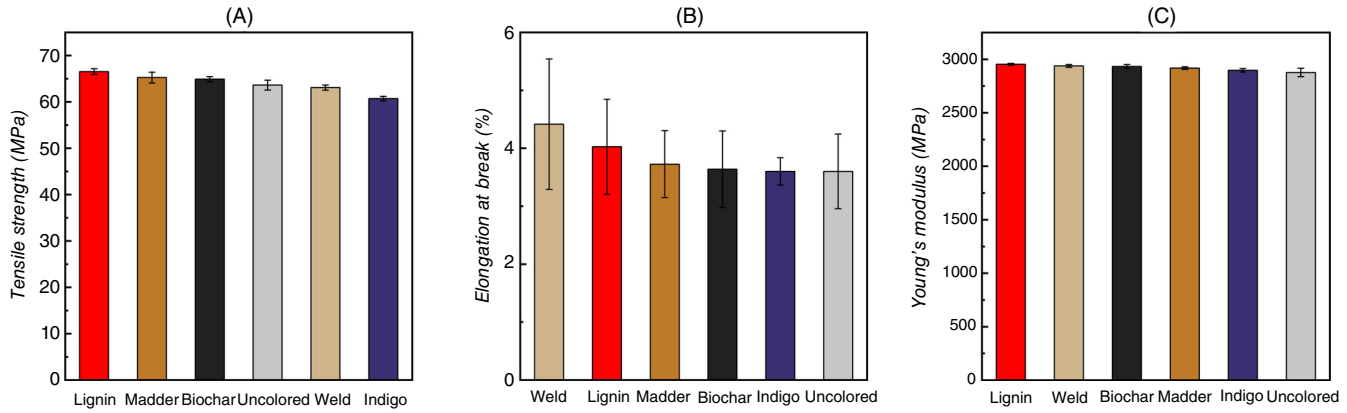


FIGURE 5 Tensile properties of the plastics. (A) Average ultimate tensile strength, (B) average elongation at break, and (C) Young's modulus. The error bars represent 95% confidence interval for a single measurement. Colours in the figure: Black = biochar, blue = indigo, red = lignin, bronze = madder, ecru = weld, and grey = uncoloured polylactic acid.

properties, they may act as reinforcing additives for PLA. For instance, a previous study has reported that weak bonding between PLA and the rigid polyphenol network of lignin may create a load-transferring interface, thus enhancing mechanical properties of the plastic.[69] Contrary to the other coloured plastics, the indigo- and weld-coloured plastics both achieved lower tensile strength than uncoloured PLA. However, the difference between weld-coloured and uncoloured plastic was not statistically significant. Indigo-coloured plastic, however, was the weakest one by a substantial margin, which hints that indigo might weaken the tensile properties of injection moulded PLA.

Relative elongation at break of the plastics during tensile test is presented in Figure 5B and their Young's moduli in Figure 5C. Figure 5B shows that there were no statistically significant variations in elongation between the plastics, even though the weld-coloured specimens appeared to strain slightly more than the other specimens. Interestingly, the uncoloured and weld-coloured specimens presented larger deviation in their elongation properties compared to the other specimens. Nevertheless, for individual specimens, no correlation between tensile strength and elongation at break was observed. Figure 5C, in turn, shows that all colourants seemed to slightly increase the Young's modulus of PLA. Despite the minimal differences in average Young's modulus, a statistically relevant difference between the lignin-coloured PLA (2953 ± 8 MPa) and the uncoloured PLA (2876 ± 37 MPa) was observed, probably due to the previously described lignin–PLA interaction.

4 | CONCLUSIONS

The present study showed that an injection moulding process could be used to manufacture biocoloured PLA with desirable visual appearance and good plastic quality. Excellent to moderate lightfastness properties were achieved with all biocolourants except indigo, which suffered from particle agglomeration during injection moulding. Furthermore, all biocolourants provided substantial protection for PLA against photodegradation. The best protection against photodegradation was achieved with lignin, which fully prevented the photo-oxidative reaction during xenon light exposure. In addition to providing chemical protection, lignin also improved tensile strength of PLA. Besides lignin, madder extract and biochar improved tensile properties of the plastic as well.

Based on the results of visual, spectral, chemical, and mechanical investigations conducted, kraft lignin can be viewed as a highly promising, protective, and strengthening biocolourant additive for injection moulded PLA products. Madder extract and biochar also showed promising results, even though PLA coloured with madder extract presented only moderate colour fastness properties and the colour achieved with biochar was not as uniform as the colours achieved with madder extract or lignin. In contrast to the desirable results with other colourants, PLA coloured with weld extract showed poor colour strength and PLA coloured with natural indigo presented an uneven colour due to agglomeration of the colourant particles. In summary, further studies on lignin–PLA interactions and more practical trials on kraft lignin as PLA additive are highly warranted, while more fundamental approach is probably better suited for the other colourants.

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