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Sustainability Spotlight

View Article Online
DOI: 10.1039/D6SU00184J

This research exemplifies a systems-level approach to sustainable polymer development, integrating life cycle assessment, synthesis optimisation, and process modelling to guide material innovation. By coupling bio-derived monomers and crosslinkers with degradable polymer architectures, the work supports a transition from linear to circular composite manufacturing. The demonstrated improvements in environmental performance across multiple impact categories highlight the value of quantitative sustainability assessments in materials design. Overall, the study provides both a methodological template and a practical route for reducing resource dependence and waste generation in thermosetting resin production.

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DOI: 10.1039/D6SU00184J

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ARTICLE

Life Cycle Assessment of Thermosets Built from a Biobased Hardener

Received 00th January 20xx,
Accepted 00th January 20xx

DOI: 10.1039/x0xx00000x

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Epoxy resins are among the most widely used matrices in fibre-reinforced polymer composites (FRPs) because their crosslinked networks provide the stiffness, strength, and thermal stability required for high-performance structural applications. As both the embodied fibres and chemical constituents have intrinsically high environmental impacts, circularity and sourcing can improve product sustainability. However, crosslinks prevent remelting or remoulding of the materials once cured, meaning that conventional mechanical recycling and reprocessing routes are not possible. Moreover, mechanical recycling can damage fibre reinforcements, reducing both their mechanical performance and economic value. As a result, FRP recycling must rely on chemical methods capable of selectively depolymerising the networks, whilst leaving the fibre reinforcements intact. We showed that replacing conventional epoxy cross-linkers (hardeners) with a biobased alternative bis(1,3-dioxolan-4-one) (bisDOX) can enable circularity. Previously, we demonstrated the use of bisDOX as a drop-in replacement for epoxy thermosets and in fully depolymerisable polyester FRPs. Herein, we seek to optimise these systems and quantify the environmental impacts compared to conventional alternatives. Life cycle assessments (LCA) identified hot spots in the synthesis of bisDOX, with process optimisation dramatically reducing its environmental impacts by up to 60%. Finally, through construction of a process model, we demonstrate the potential for these impacts to be reduced even further, by up to 70% compared to the original lab-scale synthesis, providing further sustainability arguments for modernising both epoxy and ester FRP designs.

Sustainability Spotlight

This research exemplifies a systems-level approach to sustainable polymer development, integrating life cycle assessment, synthesis optimisation, and process modelling to guide material innovation. By coupling bio-derived monomers and crosslinkers with degradable polymer architectures, the work supports a transition from linear to circular composite manufacturing. The demonstrated improvements in environmental performance across multiple impact categories highlight the value of quantitative sustainability assessments in materials design. Overall, the study provides both a methodological template and a practical route for reducing resource dependence and waste generation in thermosetting resin production.

1. Introduction

Plastics comprise the clothes we wear, the cars we drive, the places we stay and the sports we play. They package the medicines we take, the food we eat, and the way we transport goods. Thermosets, used when performance requirements are high, comprise cross-linked polymer chains and account for 15-20% of total plastic production.[1] Their excellent thermomechanical properties making them useful for structural applications,[2] particularly when reinforced with fibres such as glass and carbon to create fibre reinforced polymer composites (FRPs).[3] In particular, epoxy resins are popular matrix materials for FRPs, due to their excellent heat and chemical resistance, adhesion strength, tensile strength, rigidity and toughness.[4] Epoxy resins are synthesised from an epoxy monomer, a hardener and, sometimes, a catalyst. Their cross-linked network structure precludes recyclability, limiting their end-of-life options to landfill and incineration, both of which have significant environmental implications.[5] Furthermore, currently, epoxy monomers and amine hardeners are synthesised through environmentally impactful processes using non-renewable, petroleum derived feedstocks,[6,7] which further impedes their sustainability.

One of the major applications of epoxy-based FRPs is in the manufacture of wind turbine blades, with sharp rises in both number and size[8] at odds with the escalating epoxy-based composite waste produced, with up to up to 43 million tonnes of decommissioned wind turbine blades will accumulate worldwide by 2050.[9] Consequently, there is a strong drive to develop next-generation epoxy systems that combine excellent mechanical and thermal performance with improved environmental sustainability. Ideally, these materials would be both recyclable and incorporate monomers and hardeners with lower environmental footprints.

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DOI: 10.1039/D6SU00184J

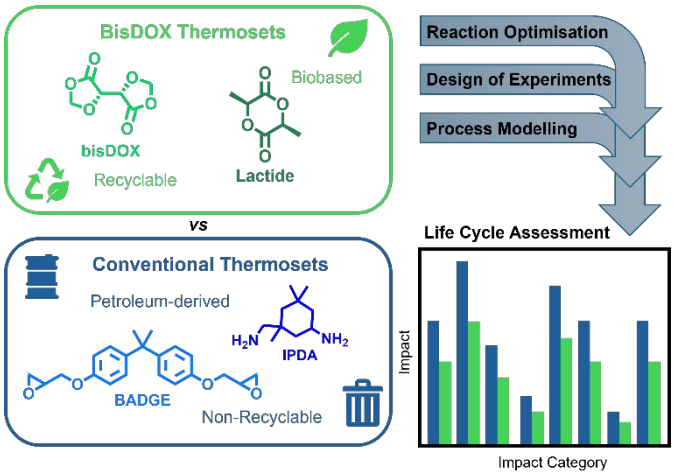

Figure 1: Schematic depicting structures of the constituents of bisDOX thermosets compared with conventional thermosets, and the methodologies employed to improve the environmental impacts associated with the synthesis of bisDOX thermosets

To avoid greenwashing, quantifying comparative environmental impacts is essential. In this paper we use life cycle assessment (LCA) to examine the environmental impacts associated with both materials and processes,[10] comparing impacts associated with the synthesis of thermosetting polymers made from i) a petroleum-derived monomer and a petroleum-derived hardener, ii) a petroleum-derived monomer and a biobased hardener and iii) a biobased monomer and a biobased hardener (**Figure 1**). The petroleum derived monomer and hardener used were bisphenol-A derived diglycidyl ether (BADGE) and isophorone diamine (IPDA), respectively, as they are widely used industrially.[11,12] Our group has previously reported that bis(1,3-dioxolan-4-one) (bisDOX),[13,14] produced from L-(+)-tartaric acid as a by-product of wine production,[15] is an efficient biobased hardener. Importantly, bisDOX thermosets are also recyclable, which further strengthens the environmental argument for their use. Although bisDOX-hardened thermosets meet demanding performance requirements, one should not assume a reduced environmental impact compared with conventional resins. Thus, this work aims to explore their sustainability credentials through use of LCA, improve these credentials using reaction optimisation to address hot spots, and finally investigate the industrial viability of their production through process modelling.

2. Results and Discussion

LCA Comparison of bisDOX Thermosets and Conventional Thermosets

The most widely used epoxy hardeners are petroleum-derived amines such as isophorone diamine (IPDA), meta-xylene diamine (*m*-XDA) and triethylenetetramine (TETA) (**Figure S1**).[12,16,17] LCA was used to determine whether the use of alternative hardener bisDOX could reduce the environmental impacts associated with FRP production. Since IPDA was the most frequently encountered hardener in the literature,[12] it was chosen as the comparator. Moreover, a preliminary LCA conducted comparing the impacts of producing IPDA, *m*-XDA, and TETA showed that IPDA had the lowest environmental impacts among the three hardeners (**Table S17**). Consequently, if bisDOX demonstrated lower impacts than IPDA, it could be reasonably inferred that its impacts would also be lower than those of *m*-XDA and TETA, thus only IPDA was included in further assessments. The goal of this study was to assess the environmental impacts associated with the synthesis of a) BADGE resins hardened with bisDOX and b) BADGE resins hardened with IPDA, both of which have been described in our previous work.[18] In this LCA, bisDOX is referred to as bisDOX-A, as it is the first iteration of the synthetic methodology to prepare the cross-linker; bisDOX-B and bisDOX-C will be introduced later on (**Table 1**). The scope was a cradle-to-gate approach, covering all inputs (materials and energy) involved in the synthesis of the resins (**Figure 2a**), and the functional unit (FU) was defined as *"the synthesis of 1 kg of resin"*. The system boundary encompassed synthesis of the monomer, hardener, and final resin (*e.g.*, bisDOX-A and BADGE production followed by curing to form **BADGE-bisDOX-A**). The environmental impacts were assessed using the ReCiPe 2016 Midpoint impact assessment method across the categories listed in **Table 2**. Transport, product manufacture, use phase, and end-of-life were excluded, as these were assumed to be comparable across resin systems with similar applications and lifespans. This approach isolates the impact of feedstock origin and resin synthesis, avoiding uncertainty associated with context-dependent disposal and recycling scenarios, which are often speculative and poorly defined at the material development

Table 1: Nomenclature used to describe the three iterations of bisDOX.

Name	Definition
bisDOX-A	Original
bisDOX-B	Post-optimisation
bisDOX-C	Post-scale-up modelling

Table 2: Environmental impact categories used for the assessment of resin syntheses.

Impact Category	Units
Global warming,	kg CO_2 eq.
Stratospheric ozone depletion	kg CFC11 eq.
Ionizing radiation,	kBq Co-60 eq.
Ozone formation (human health	kg NO_x eq.
Ozone formation (terrestrial ecosystems)	kg PM2.5 eq.
Fine particulate matter formation	kg NO_x eq.
Terrestrial acidification	kg SO_2 eq.
Marine eutrophication	kg P eq.
Freshwater eutrophication	kg N eq.
Marine ecotoxicity	kg 1,4-DCB
Freshwater ecotoxicity	kg 1,4-DCB
Terrestrial ecotoxicity	kg 1,4-DCB
Human carcinogenic toxicity	kg 1,4-DCB
Human non-carcinogenic toxicity	kg 1,4-DCB
Land use	m2a crop eq.
Mineral resource scarcity	kg Cu eq.
Fossil resource scarcity	kg oil eq.
Water consumption	m^3

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stage and may unfairly bias towards the recyclable bisDOX system if carbon fibre recovery is presumed rather than realised.

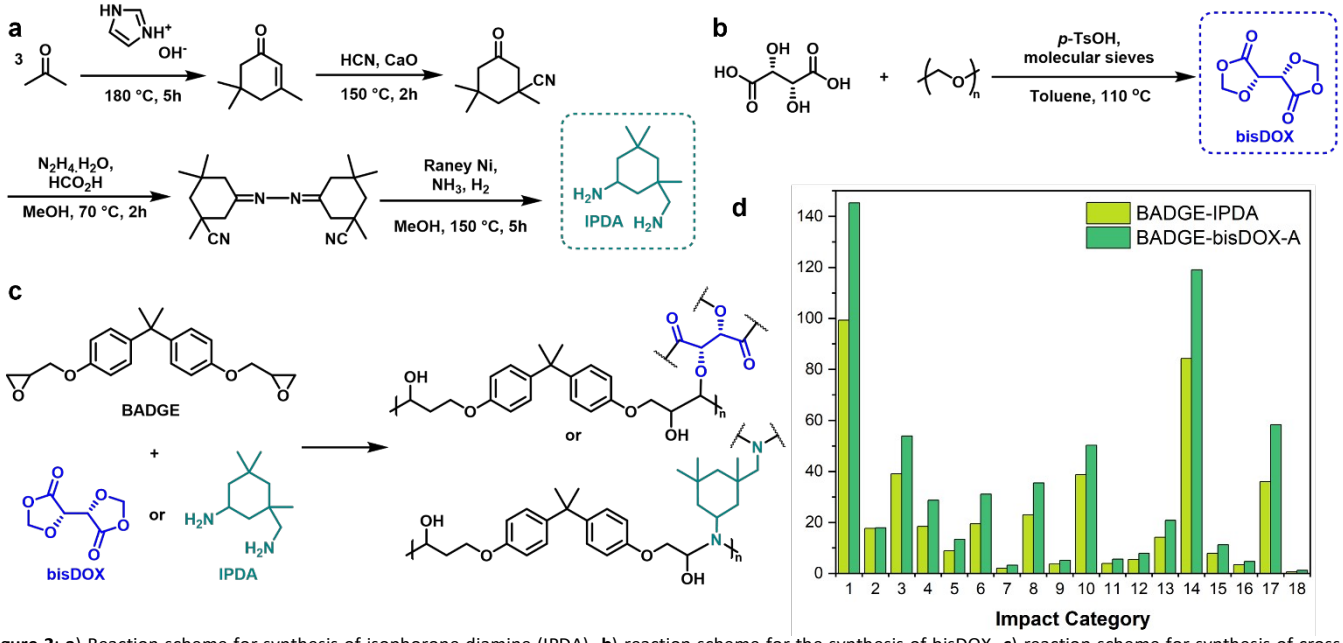

Figure 2: **a**) Reaction scheme for synthesis of isophorone diamine (IPDA), **b**) reaction scheme for the synthesis of bisDOX, **c**) reaction scheme for synthesis of cross-linked epoxy resins from bisphenol-A diglycidyl ether (BADGE) monomer with bisDOX or isophorone diamine (IPDA) hardener, and **d**) LCA comparison of BADGE hardened with IPDA vs bisDOX-A (unoptimised). Impact categories: 1) Global warming, kg CO_2 eq., 2) Stratospheric ozone depletion, kg CFC11 eq.×10^4, 3) Ionizing radiation, kBq Co-60 eq., 4) Ozone formation, Human health, kg NOx eq.×10^2, 5) Fine particulate matter formation, kg PM2.5 eq.×10^2, 6) Ozone formation, Terrestrial ecosystems, kg NOx eq.×10^2, 7) Terrestrial acidification, kg SO_2 eq.×10, 8) Freshwater eutrophication, kg P eq.×10^3, 9) Marine eutrophication, kg N eq.×10^3,10) Terrestrial ecotoxicity, kg 1,4-DCB/10, 11) Freshwater ecotoxicity, kg 1,4-DCB, 12) Marine ecotoxicity, kg 1,4-DCB, 13) Human carcinogenic toxicity, kg 1,4-DCB, 14) Human non-carcinogenic toxicity, kg 1,4-DCB, 15) Land use, m^2a crop eq., 16) Mineral resource scarcity, kg Cu eq.×10, 17) Fossil resource scarcity, kg oil eq., 18) Water consumption, m^3.

The analysis showed that biobased bisDOX had higher impacts in 16 of the 18 assessed categories (**Figure 2d**). Notably, **BADGE-bisDOX-A** showed a 30% increase in global warming potential, indicating that the synthesis of bisDOX-A had higher greenhouse missions compared to the synthesis of IPDA. In the other categories, impact increases ranged from relatively minor (1% for stratospheric ozone depletion) to more pronounced (up to 89% for water consumption). This outcome was anticipated to some extent, as bisDOX was synthesised in-house using a laboratory-scale, non-optimised process (**Figure 2b**), whereas IPDA is produced at industrial scale *via* a well-established and highly optimised synthetic route (**Figure 2a**). Studies have consistently shown that industrial-scale chemical processes tend to exhibit significantly lower environmental impacts than their laboratory-scale counterparts, primarily due to enhanced process efficiency and resource utilisation.[19] Thus, we decided to optimise the bisDOX synthesis procedure, to explore whether this would reduce its impacts and how this would impact the LCA results.

Process Optimisation of bisDOX Synthesis

The initial LCA of bisDOX synthesis (**Figure 2d**) prompted an investigation into hot spots in the process that could be contributing to its high impacts. These factors identified included: a long reaction time at high temperature (16 h at 110 °C), a large number and quantity of solvents being used (toluene, EtOAc, hexane and THF) and a low isolated yield of product (20-30%) being obtained. Hence, process optimisation of the synthetic route was carried out, to reduce its environmental impacts. The first stage of optimisation entailed screening multiple catalysts, solvents and atmospheres (**Figures S7-9**), to see which conditions afforded bisDOX in the highest yields. Reaction conversions were monitored by ^{1}H NMR, but because the starting material (L-(+)-tartaric acid) was sparingly soluble, accurate conversions could not be calculated by comparing ratios of reactants to products. Instead, conversions were calculated by using diphenyl ether (DPE) as an external standard (**Supplementary Information, Section 1.2**). This screening found the optimal solvent to be toluene and catalyst to be *p*-TsOH, but that atmosphere did not have a significant impact on the yield of product obtained. Attempts to improve yields through catalyst and solvent variations proved unsuccessful, however through monitoring conversion over time using NMR, a vital observation was made. The yield of bisDOX increased from 0-5 h but then decreased from 5-16 h. Thus, the low yields obtained previously could be attributed to the use of an excessive 16 h reaction time. By reducing the time, not only would the yield of bisDOX increase, but the energy requirements for the process would vastly decrease, thereby greatly reducing the environmental impacts of the synthesis. However, although the NMR yield of bisDOX was now up to 80%, the isolated yield still remained a paltry 30%. This large disparity is indicative of an instability of the bisDOX during the work-up procedure. Thus, optimisation of this stage of the process targeted a reduction in losses during work-up and purification. To identify when bisDOX was lost, conversion NMR was carried out after each stage of work-up (**Figure S10**). The first key finding was that after work-up, prior to purification, the bisDOX

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obtained was pure (by NMR), negating the need for the precipitation step.

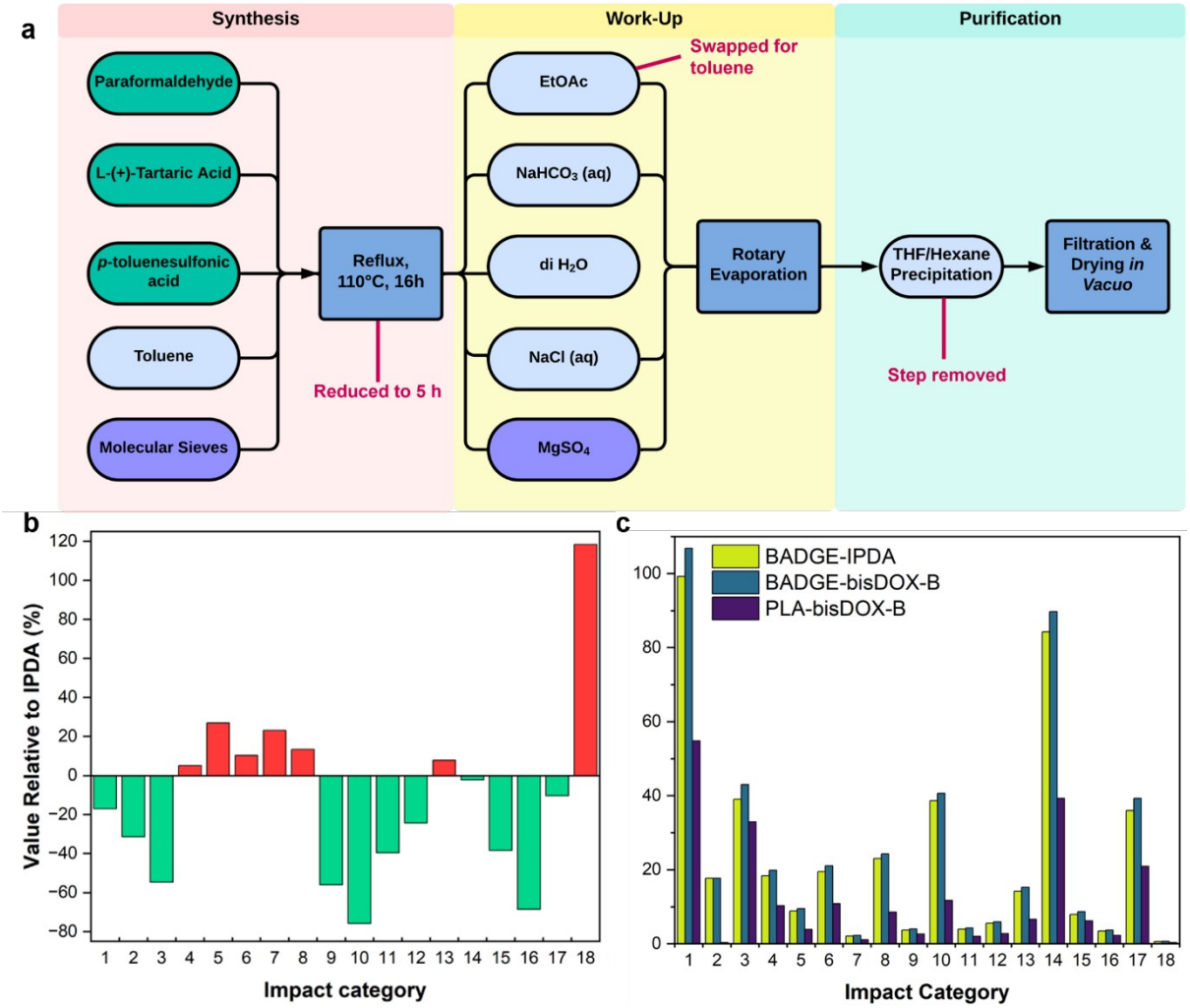

Figure 3: **a**) Flow diagram illustrating the original procedure for bisDOX synthesis, work-up and purification, highlighting changes made during process optimisation **b**) Value of bisDOX-B impacts compared with IPDA, and **c**)) LCA comparison of **BADGE-IPDA**, **BADGE-bisDOX-B** and **PLA- bisDOX-B** Impact categories: 1) Global warming, kg CO_2 eq., 2) Stratospheric ozone depletion, kg CFC11 eq.×10^4, 3) Ionizing radiation, kBq Co-60 eq., 4) Ozone formation, Human health, kg NOx eq.×10^2, 5) Fine particulate matter formation, kg PM2.5 eq.×10^2, 6) Ozone formation, Terrestrial ecosystems, kg NOx eq.×10^2, 7) Terrestrial acidification, kg SO_2 eq.×10, 8) Freshwater eutrophication, kg P eq.×10^3, 9) Marine eutrophication, kg N eq.×10^3,10) Terrestrial ecotoxicity, kg 1,4-DCB/10, 11) Freshwater ecotoxicity, kg 1,4-DCB, 12) Marine ecotoxicity, kg 1,4-DCB, 13) Human carcinogenic toxicity, kg 1,4-DCB, 14) Human non-carcinogenic toxicity, kg 1,4-DCB, 15) Land use, m^2a crop eq., 16) Mineral resource scarcity, kg Cu eq.×10, 17) Fossil resource scarcity, kg oil eq., 18) Water consumption, m^3.

It was also found that the first step of work-up, dilution with EtOAc, was causing a decrease in bisDOX yield of 55%, so a variety of solvents were screened (**Figure S11**) and it was found that the use of toluene did not cause a reduction in bisDOX yield and so was more suitable. Small quantities of bisDOX were lost during subsequent aqueous washes, which was to be expected due to its hydrolytic instability. The formation of bisDOX is a reversible reaction, which produces water in the forward direction, so the introduction of water causes equilibrium to shift to the left, reducing bisDOX yield. However, even with these losses, bisDOX synthesised under the optimised conditions (**Supplementary Information, Section 1.2**), named bisDOX-B, was obtained in an isolated yield of 50%, which was a two-fold increase on the pre-optimisation yields. A second LCA, using the same methodology, compared the synthesis of bisDOX-B with that of IPDA. In this case, bisDOX exhibited lower impacts in 11 of the 18 categories (**Figure 3b**). The largest reductions were observed for ionising radiation (55%), marine eutrophication (56%), terrestrial ecotoxicity (76%), and mineral resource scarcity (69%). These substantial improvements across multiple categories demonstrate the effectiveness of the reaction optimisation in enhancing the environmental performance of bisDOX synthesis, reinforcing its potential as a more sustainable epoxy hardener.

LCA and Mechanical Property Comparison of Petroleum-Based, Partially Biobased and Fully Biobased Thermosets

After optimisation of bisDOX synthesis, an LCA was carried out to compare the impacts of the production of BADGE resins with bisDOX-B *vs* IPDA as hardener, and it was observed that bisDOX hardened resins had lower impacts in 14 of the 18 categories (**Figure 3c**). However, the system could further be improved by swapping from BADGE to a biobased monomer. We have

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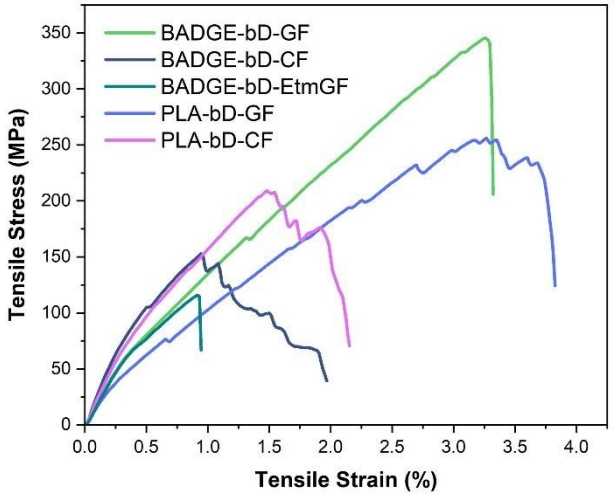
previously reported the synthesis of poly(lactic acid) (PLA) resins, cross-linked with bisDOX.[13,14] PLA is synthesised from L-(+)-lactide, a biobased monomer derived from fermentation of sugars from plants such as sugarcane and corn.[20]

Table 3: Mechanical properties of fibre reinforced BADGE and PLA composites. [a]σ_b (stress at break), ε_b (strain at break) and E (Young's modulus) data obtained from tensile testing measurements, [b]Fibre content obtained from TGA analyses.

Sample	σ_b (MPa)[a]	ε_b (%)[a]	*E* (GPa)[a]	Fibre Content (%)[b]
BADGE-bD-GF	289 ± 31	3 ± 0	18 ± 1	57 ± 1
BADGE-bD-CF	119 ± 17	1 ± 0	28 ± 2	58 ± 1
PLA-bD-GF	197 ± 27	2 ± 0	16 ± 2	71 ± 2
PLA-bD-CF	63 ± 14	2 ± 1	18 ± 3	59 ± 2

Figure 4: Tensile testing plots for BADGE and PLA fibre reinforced composites. Tensile testing measurements were conducted on a static testing Instron fitted with a 2kN load cell, at constant speeds of 200 mm/min.



An LCA was carried out to compare fully biobased **PLA-bisDOX-B** resins to partially biobased **BADGE-bisDOX-B** resins using a functional unit of 1kg of resin. PLA resins had lower impacts in every category, making them a clear front-runner in terms of environmental sustainability (**Figure 3c**). Commercially, epoxy matrices are employed over polyesters in FRPs used for high performance applications due to their superior mechanical properties.[14] In our previous work, it was found that fibre reinforced PLA composites had excellent mechanical properties that matched BADGE composites,[18] but it is important to assess the relevance and limitations of functional unit choices. The properties of **PLA-bisDOX-GF/CF** were compared with those of **BADGE-bisDOX-GF/CF** composites (**Table 3, Figure 4**). While **BADGE-bisDOX-GF** had a slightly higher tensile stress at break (σ_b) and elongation at break (ε_b) than **PLA-bisDOX-GF**, their Young's moduli (*E*) were not significantly different. Meanwhile, **BADGE-bisDOX-CF** had a higher σ_b and *E* than **PLA-bisDOX-CF**, but a lower ε_b. As their properties were within the same order of magnitude, performance is not sacrificed in the name of sustainability. We recognise that other parameters may be important in defining the functional unit for a specific application space.

Of note, previous work on bisDOX-epoxy FRPs highlighted processability challenges from the insolubility of bisDOX in liquid epoxy monomers, which hindered resin flow through a vacuum assisted resin infusion (VARI) set-up.[18] Thus, we explored the use of reactive diluents to improve the processability of bisDOX resins, and to minimise the experimental load associated with formulation development, design of experiments (DoE) methodology was employed (full details in **Supplementary Information, Section 1.1**). However, although dilution of the resin mixture improved its processability, the resultant FRPs had poorer mechanical properties than bisDOX-epoxy FRPs, thus we looked at alternative ways of improving the resins.

Process Modelling of bisDOX Synthesis

Although bisDOX-B now had lower environmental impacts than IPDA, opportunities for further improvement remained. Studies have shown that industrial-scale processes generally have lower impacts than lab-scale[19], so it was highly likely that scaling up bisDOX synthesis would further reduce its impacts. A process model (**Figure 5a**) for the production of bisDOX on an industrial scale (bisDOX-C) was created using the Aspen Plus software package. In this model, L-(+)-tartaric acid, paraformaldehyde, and *p*-TsOH (in a 1:4:0.2 molar ratio) were mixed with toluene and pumped through a heater into a stirred batch reactor at a flow rate of 4000 kg h^{-1}. The reaction proceeded at 110 °C and 1 bar for 5 h. This input rate was selected as it is comparable with the industrial-scale production of speciality chemicals,[21] while the reaction conditions correspond to those identified during bisDOX synthesis optimisation (**Supplementary Information, Section 1.2**). After completion, the crude reaction mixture was cooled to 25 °C and subjected to three sequential washing and phase separation steps in column separators operating at 25 °C and 1 bar, with the aqueous phase removed after each wash. After the final wash, the remaining organic phase is pumped into a stirred mixer, where it was treated with $MgSO_4$ to remove residual moisture. The mixture was then subjected to solid-liquid filtration to remove $MgSO_4$, and the filtrate was dried under a reduced pressure of 0 bar to eliminate any remaining toluene. The process yielded pure, dry bisDOX at a production rate of 1 t h^{-1}, with an overall energy requirement of 246 kcal kg^{-1}, which is equivalent to 0.286 kWh.

The model was constructed with several assumptions; however, it provides a practical approximation for the process design. The reaction between L-(+)-tartaric acid, paraformaldehyde, and *p*-TsOH in toluene is assumed to proceed to complete conversion under the specified conditions, neglecting side reactions and by-product formation. All liquid phases are treated as ideal, and phase separations during sequential washing steps are assumed to be complete, with no loss of bisDOX to the aqueous fractions.

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The $MgSO_4$ and oven drying steps are assumed to remove all residual toluene without affecting product purity. Scale-up from laboratory to industrial production is assumed linear, maintaining reaction kinetics and separation efficiencies, while potential operational inefficiencies, including incomplete mixing, equipment limitations, heat losses, and mass transfer constraints are not accounted for.

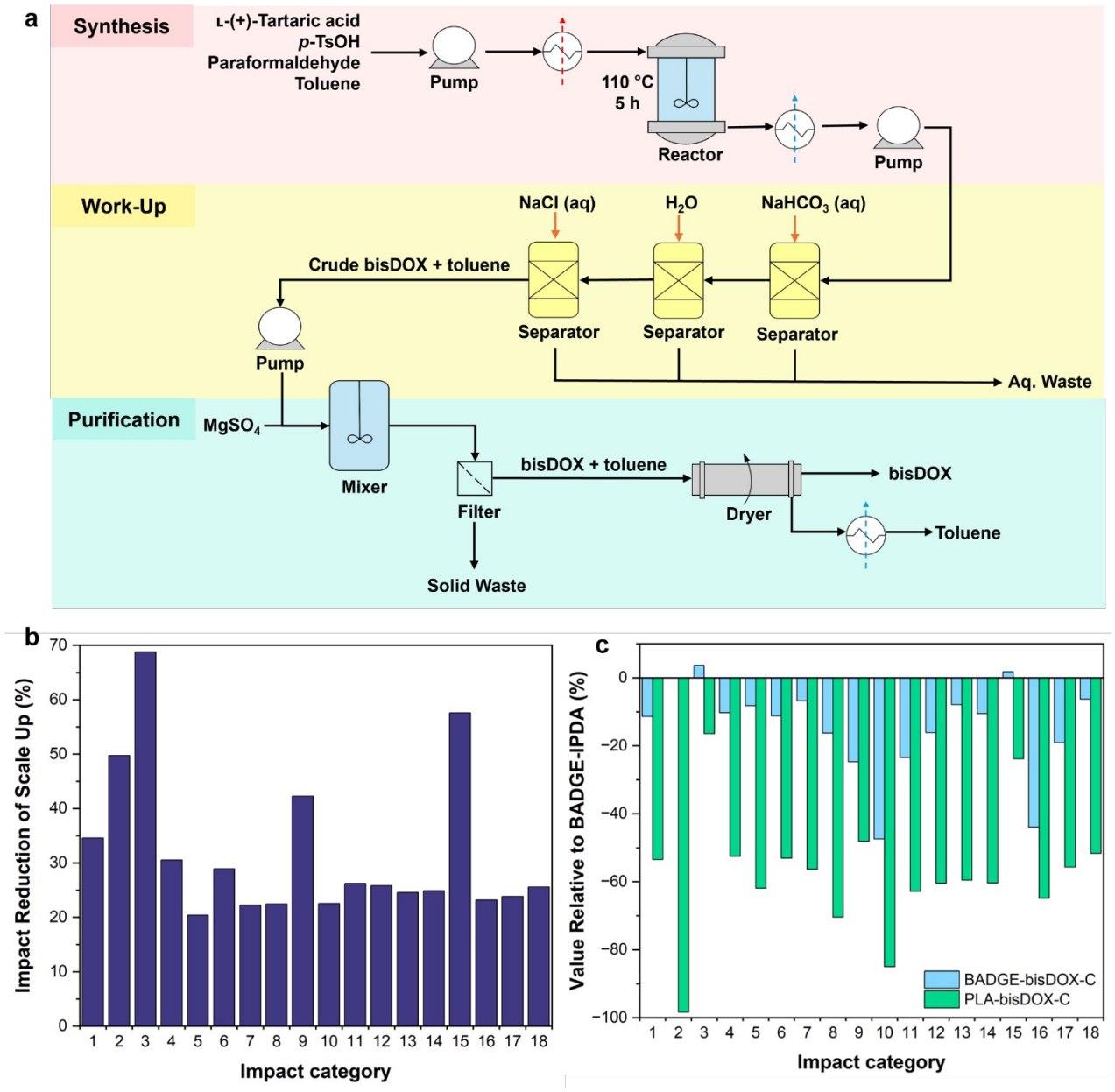

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Figure 5: **a**) Process flow diagram for bisDOX synthesis, **b**) Reduction in environmental impacts when bisDOX is synthesised on an industrial-scale compared to lab-scale, and **c**) Impacts of BADGE-bisDOX-C and PLA-bisDOX-C compared with BADGE-IPDA synthesis. Impact categories: 1) Global warming, kg CO_2 eq., 2) Stratospheric ozone depletion, kg CFC11 eq.×10^4, 3) Ionizing radiation, kBq Co-60 eq., 4) Ozone formation, Human health, kg NOx eq.×10^2, 5) Fine particulate matter formation, kg PM2.5 eq.×10^2, 6) Ozone formation, Terrestrial ecosystems, kg NOx eq.×10^2, 7) Terrestrial acidification, kg SO_2 eq.×10, 8) Freshwater eutrophication, kg P eq.×10^3, 9) Marine eutrophication, kg N eq.×10^3,10) Terrestrial ecotoxicity, kg 1,4-DCB/10, 11) Freshwater ecotoxicity, kg 1,4-DCB, 12) Marine ecotoxicity, kg 1,4-DCB, 13) Human carcinogenic toxicity, kg 1,4-DCB, 14) Human non-carcinogenic toxicity, kg 1,4-DCB, 15) Land use, m^2a crop eq., 16) Mineral resource scarcity, kg Cu eq.×10, 17) Fossil resource scarcity, kg oil eq., 18) Water consumption, m^3.

Solvent recovery was excluded from the process model, representing a conservative assumption. Incorporating solvent recovery in a future, optimised model would likely further reduce the environmental impacts of the process. The material and energy requirements from the process model were extracted and inputted into a final LCA (full inventory in **Supplementary Information, Section 2**), and the results showed that the impacts in every category were reduced by 20-70% compared to the original lab-scale synthesis (**Figure 5b**). A direct comparison of **BADGE-IPDA** and **BADGE-bisDOX-C** production (**Figure 5c**) further revealed that **BADGE-bisDOX-C** had lower impacts in 16 of 18 categories. To evaluate the robustness of these results and reduce uncertainty associated with proxy selection in the life cycle inventory (**Supplementary Information, Section 2**), a sensitivity analysis was performed. Increasing the mass of each chemical proxy by 20% to simulate a worst-case scenario did not affect the relative ranking of impact categories within each resin system, nor the comparative ordering of the three resins. This indicates that the principal conclusions are not sensitive to plausible systematic errors in the inventory data. A second sensitivity analysis was conducted to evaluate the influence of assumptions made during construction of the process model, specifically operational inefficiencies, including incomplete mixing, heat losses, and mass transfer constraints, all of which would result in a reduction in bisDOX yield relative to the modelled value of 50%. Decreasing the isolated yield of bisDOX from 50% to 40% did not affect the relative ranking of impact categories within or between resin systems, which indicted that the modelling assumptions were robust with respect to the comparative conclusions within the system boundary. Furthermore, we have previously demonstrated the recyclability of FRPs with both **PLA-bisDOX**[13,14] and **BADGE-bisDOX**[18] matrices, so together with their lower production impacts relative to **BADGE-IPDA**, this highlights their potential to deliver improved sustainability across both manufacturing and end-of-life stages, although this lies outside the current system boundary and is therefore not captured in the LCA results.

3. Conclusions

Herein, we have demonstrated that biobased feedstocks alone do not inherently guarantee lower environmental impacts; rather, efficient synthesis routes and scalable manufacturing processes are critical to achieving genuinely sustainable thermosetting resin systems. By integrating LCA, reaction optimisation, and process modelling, we developed bisDOX-hardened resins with substantially improved sustainability credentials relative to conventional IPDA-hardened systems. Initial LCA results showed that **BADGE-bisDOX** had higher environmental impacts than **BADGE-IPDA** across all categories, despite the biobased content. After optimising bisDOX synthesis at laboratory scale and modelling its industrial-scale production, the projected process exhibited lower impacts than IPDA in 16 of 18 categories, with an average 14% reduction across all impact categories. In addition, bisDOX is bio-based, reducing reliance on fossil resources, and introduces labile ester linkages that promote resin degradability and reduce non-recyclable waste.

We have also established that the impacts of thermosetting resin production can be further reduced when both the monomer and cross-linker are substituted for biobased alternatives. This was achieved by carrying out an LCA to compare the syntheses of **BADGE-IPDA** (fully petroleum-based), **BADGE-bisDOX** (partially biobased) and **PLA-bisDOX** (fully biobased). The data showed that **PLA-bisDOX** had by far the lowest impacts in every category, with an average 57% reduction across all impact categories compared with **BADGE-IPDA**. Moreover, unlike **BADGE-IPDA**, **PLA-bisDOX** and its FRPs are fully recyclable.[14] Collectively, these findings highlight that meaningful reductions in the environmental footprint of thermosetting resins require not only renewable feedstocks, but also optimised and industrially viable production strategies capable of supporting a transition from a linear plastics economy to a circular one. It is important to note that these environmental improvements should be considered at the level of the complete resin formulation and its production, rather than attributing the benefit to any component in isolation.

Author contributions

Eloise Billington: Conceptualization, Data Curation, Formal analysis, Investigation Methodology, Software, Validation, Writing – Original draft, Writing – Review & Editing
Michael P Shaver: Supervision, Funding acquisition, Writing – Review & Editing.
Ciaran W Lahive: Writing – Review & Editing.

Funding

This work was conducted with financial support from the EPSRC (Grant EP/S025200/1), the Henry Royce Institute for Advanced Materials (Grants EP/R00661X/1, EP/S019367/1, EP/P025021/1, and EP/P025498/1), and the European Regional Development Fund OC15R19P 0903.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the Supplementary Information.

Acknowledgements

The authors would like to thank Dr Wanrong Wang for helpful advice regarding process modelling.

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View Article Online
DOI: 10.1039/D6SU00184J

The data supporting this article have been included as part of the Supplementary Information