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Sulfonic acid group functionalized Brönsted acidic ionic liquid catalyzed depolymerization of *poly*(ethylene terephthalate) in water

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ABSTRACT

Sulfonic acid group functionalized 1,3-dialkyl imidazolium Brönsted acidic ionic liquids are shown as better catalysts in comparison to sulfuric acid with similar acid concentration for the depolymerization of *poly*(ethylene terephthalate) in hot compressed water. The highest activity was found by using 1.00 M aqueous 1-(3-propylsulfonic)-3-methylimidazolium chloride catalyst, where a practically complete decomposition of *poly*(ethylene terephthalate) could be achieved after 24 h at 210 °C with a 94% isolated yield of terephthalic acid. The superior catalytic activity of 1-(alkylsulfonic)-3-methylimidazolium chloride in comparison to sulfuric acid with similar acidic strength is explained as a result of charge-transfer and dipolar interactions between catalyst and *poly*(ethylene terephthalate).

1. Introduction

The current interest in renewable resources and environmental concerns has resulted a rapid growth in research activities in many areas including, new sustainable materials (Bontempi et al., 2021), recycling of plastics and depolymerization of current polymers for reuse of feedstocks (Kosloski-Oh et al., 2021), (Miao et al., 2021), (Cho and Cho, 2020), (Sikdar, 2020) as well as biodegradable polymers (RameshKumar et al., 2020), (Rai et al., 2021), (Luckachan and Pillai, 2011). Depolymerization of used polymer materials to recover valuable monomers for reuse as chemical feedstocks is an important branch in this multipronged approach towards a sustainable future. The current global production of synthetic polymers is approximately 290 million tons per year, and the major players in this arena are polyethylene (37%), polypropylene (26%), polyvinylchloride (PVC) (18%) and polyethylene terephthalate (PET) (8%) (Delidovich et al., 2016), (Association, 2015). The vast majority of these polymers are very stable and practically impossible to depolymerize back to their monomers under mild conditions. However, among these polyethylene terephthalate has given some hope and attracted special attention; probably due to relatively labile ester link, as well as the high molecular mass, valuable and stable terephthalic acid monomer (Barnard et al., 2021), (Padhan and Sreeram, 2019), (Raheem et al., 2019).

The major chemical PET depolymerization methods includes: alcoholysis, glycolysis, hydrolysis and aminolysis (Shojaei et al., 2020). All these methods require heating at high temperatures and pressures and the first three are usually carried out in the presence of an acid, alkaline or metal based catalyst. The aminolysis is a digestion in ammonia or amine such as: methyl, ethyl, propyl, butyl, ethanol, benzyl amine or aniline, known with catalysts: zinc acetate, lead acetate or potassium sulfate (Sadeghi and Sayaf, 2012), (Hoang and Dang, 2013). In addition, Arias and Thielemans have reported a recent development of a mild depolymerization method using anhydrous sodium methoxide, converting PET into dimethyl terephthalate in 4 min at 80 °C (Rubio Arias and Thielemans, 2021). However, this technique requires careful humidity control and heating using microwaves (Rubio Arias and Thielemans, 2021). The application of ionic liquids in depolymerization is rare, and only a handful of examples are reported where ionic liquids are used as catalysts and solvents in glycolysis (Wang et al., 2009), (Yue et al., 2011), (Al-Sabagh et al., 2014), (Silva et al., 2018), (Liu et al., 2009). For example, Wang et al. reported the use 1-butyl-3-methylimidazolium chloride as the catalyst for the glycolysis of PET in ethylene glycol at 180 °C, producing *bis*(hydroxyethyl)terephthalate in 78% yield (Wang et al., 2009). Furthermore, Silva and co-workers reported the use of 1-butyl-3-methylimidazolium trichlorozincate in a similar glycolysis process (Silva et al., 2018). In addition, PET hydrolysis was possible by using ionic liquid 1-butyl-3-methylimidazolium chloride as the solvent and acid-functionalized ionic liquid 1-(3-propylsulfonic)-3-methylimidazolium hydrogen sulfate as the catalyst (Liu et al., 2009). During these experiments Liu and co-workers have reported achieving a 88% yield of terephthalic acid under optimum conditions where a 1-*n*-butyl-3-methylimidazolium chloride and water mixture in 3:2 mole ratio was used as the solvent (Liu et al., 2009).

In 2009 we developed the use of sulfonic acid group (-SO_3H) functionalized Brönsted acidic ionic liquids (BAILs) for the concurrent dissolution and hydrolysis of cellulose under mild conditions

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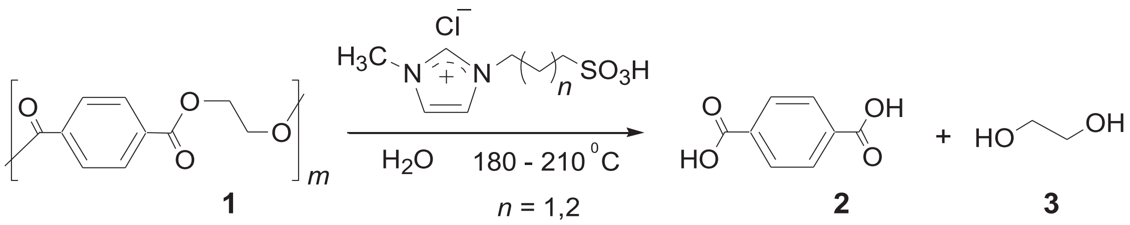

Fig. 1. Brönsted acidic ionic liquid (BAIL) catalyzed depolymerization of *poly*(ethylene terephthalate) (**1**) to terephthalic acid (**2**) and ethylene glycol (**3**) in water.

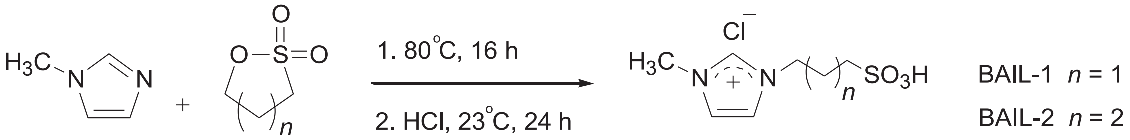

Fig. 2. Synthesis of 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) and 1-(4-butylsulfonic)-3-methylimidazolium chloride (BAIL-2) Brönsted acidic ionic liquids.

(Amarasekara and Owereh, 2009). Where we reported that high molecular weight cellulose dissolved in acidic ionic liquids 1-(3-propylsulfonic)-3-methylimidazolium chloride and 1-(4-butylsulfonic)-3-methylimidazolium chloride could be hydrolyzed at 70 °C and at atmospheric pressure in the presence of 2.0 equivalents of water per glucose unit of cellulose, to give glucose along with other reducing sugars in good yields. Later we have shown that these acidic ionic liquids can be used as homogeneous catalysts in aqueous phase as well (A.S. Amarasekara and Wiredu, 2011). For example, a dilute aqueous solution 0.0321 mol H^+/L 1-(3-propylsulfonic)-3-methylimidazolium chloride was shown to be a better catalyst than aqueous sulfuric acid of the same H^+ ion concentration for the hydrolysis of cellulose in water at 140–180 °C temperature range (A.S. Amarasekara and Wiredu, 2011). In addition, 1-(3-alkylsulfonic)-3-methylimidazolium chloride Brönsted acidic ionic liquids have shown exceptional acid catalysis activities in a number of reactions such as: Skraup synthesis of quinolines under microwave heating (Amarasekara and Hasan, 2014), dehydration of D-glucose to 5-hydroxymethylfurfural (Amarasekara and Razzaq, 2014), and oxidation of furan aldehydes with hydrogen peroxide (Amarasekara and Okorie, 2018).

In continuation of our interest in catalysis activity of sulfonic acid group functionalized Brönsted acidic ionic liquids (Amarasekara, 2016), (Amarasekara and Gutierrez Reyes, 2019), we have studied the possibility of using BAILs as catalysts for depolymerization of *poly*(ethylene terephthalate) in water. In this communication we report the first application of 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) and 1-(4-butylsulfonic)-3-methylimidazolium chloride (BAIL-2) as catalysts for the depolymerization of *poly*(ethylene terephthalate) (1) in water as shown in Fig. 1, and their superior activity in comparison to sulfuric acid.

2. Experimental

2.1. Materials and Instrumentation

1-Methylimidazole, 1,3-propanesultone, 1,4-butanesultone, hydrochloric acid (37%), sodium hydroxide were purchased from Aldrich Chemical Co. Polyethylene terephthalate (PET) was obtained from used Kirkland® 16.9 Fl OZ purified water bottles. Dry water bottles were shredded with a coarse file, sieved through a brass mesh (Aldrich mini-sieve set, Z 675,415, US mesh size 18, particle size 1.0 mm) and the PET powder was stored in a sealed plastic bottle until used for experiments. 1-(3-Propylsulfonic)-3-methylimidazolium chloride (BAIL-1) and 1-(4-butylsulfonic)-3-methylimidazolium chloride (BAIL-2) catalysts were prepared according to the literature procedures as shown in Fig. 2. (Gui et al., 2004), (Yang et al., 2008). FT-IR and ^1H NMR spectra of BAIL-1 and 2 are included in the supplementary material. PET depolymerization experiments were carried out in 25 mL stainless steel solvothermal reaction kettles with Teflon inner sleeves, from Lonsino Medical Products Co. Ltd., Jingsu, China. The tightly closed reaction kettles were heated in a preheated Precision Scientific microprocessor-controlled laboratory oven (Model: WU-52,402–91) with temperature accuracy ±1 °C. Thermogravimetric (TG) analysis was carried out in air using TA instruments TGA 2050 system. The mass of the polymer used for a scan was approximately 10 mg, and Pt crucibles were used. The TG curves were recorded in 23–800 °C range using a scanning rate of 10 °C/ min, in air. FT-IR spectra were recorded on a Thermo Nicolet IR 200 using KBr pellets at 1.0 cm^{-1} resolution. ^1H NMR Spectra of BAILs were recorded in D_2O on a Varian Mercury plus spectrometer operating at 400 MHz and chemical shifts are given in ppm downfield from TMS ($\delta = 0.00$).

2.2. General procedure for the depolymerization of poly(ethylene terephthalate)

Poly(ethylene terephthalate) powder (**1**, 0.500 g, 2.38 mmol) was placed in 5.00 mL 0.05–1.0 M aqueous BAIL-1,2 or sulfuric acid solution in a 25 mL stainless steel solvothermal reaction kettle with a Teflon inner sleeve. The reaction kettle was firmly closed and heated in a thermostated oven maintained at the desired temperature for 24–168 h. Then reaction kettle was removed from the oven and cooled under running cold water to quench the reaction. The contents were basified to pH ~ 10 with drop wise addition of 1.0 M aqueous sodium hydroxide, transferred into a centrifuge tube, diluted to 10.0 mL with deionized water, and centrifuged at 1700 *g* for 6 min to precipitate the unreacted PET. The clear supernatant was transferred to a second centrifuge tube and acidified to pH ~ 1 with drop wise addition of concentrated hydrochloric acid to give terephthalic acid (TPA, **2,** Fig. 1) as a white precipitate. The white solid product was separated by centrifuging the tube at 1700 *g* for 6 min., and decanting the clear supernatant liquid. The recovered unreacted PET and isolated TPA from depolymerizations were dried in an oven at 60 °C for 24 h. and weighed. The PET conversion (%) and TPA yield (%) were calculated as follows:

$$\text{PET conversion (\%)} = \frac{0.500\ g\ -\ weight\ of\ unreacted\ PET}{0.500\ g}\ \text{X}\ 100\%$$

$$\text{TPA yield (\%)} = \frac{weight\ of\ isolated\ TPA}{0.395\ g}\ \text{X}\ 100\%$$

The catalyst, concentration, reaction conditions used, PET conversion (%) and TPA yield (%) are shown in Table 1. The isolated product was confirmed as pure terephthalic acid by comparison of IR spectrum and TGA profile of those with an authentic sample of terephthalic acid.

Table 1
The catalyst, acid concentration, reaction condition, *poly*(ethylene terephthalate) (PET) conversion and terephthalic acid (TPA) yields during the depolymerization of *poly*(ethylene terephthalate) (PET) in water. All reactions were carried out using 0.500 g of PET powder (18 mesh), 5.00 mL of aqueous catalyst solution in 25 mL stainless steel solvothermal reaction kettles with Teflon inner sleeves.

Entry	Catalyst	Concentration	Reaction condition	PET (1) Conversion (%)	TPA (2) Yield (%)
1	BAIL-1	0.05 M	210°C, 48 h.	40	33
2	BAIL-1	0.10 M	210°C, 48 h.	51	48
3	BAIL-1	0.10 M	180°C, 168 h.	55	39
4	BAIL-1	0.50 M	210°C, 24 h.	70	65
5	BAIL-1	1.00 M	210°C, 24 h.	99	94
6	BAIL-2	0.50 M	210°C, 24 h.	65	61
7	BAIL-2	1.00 M	210°C, 24 h.	92	90
8	H_2SO_4	0.05 M	210°C, 24 h.	30	18
9	H_2SO_4	0.25 M	210°C, 24 h.	56	52
10	H_2SO_4	0.50 M	210°C, 24 h.	65	63

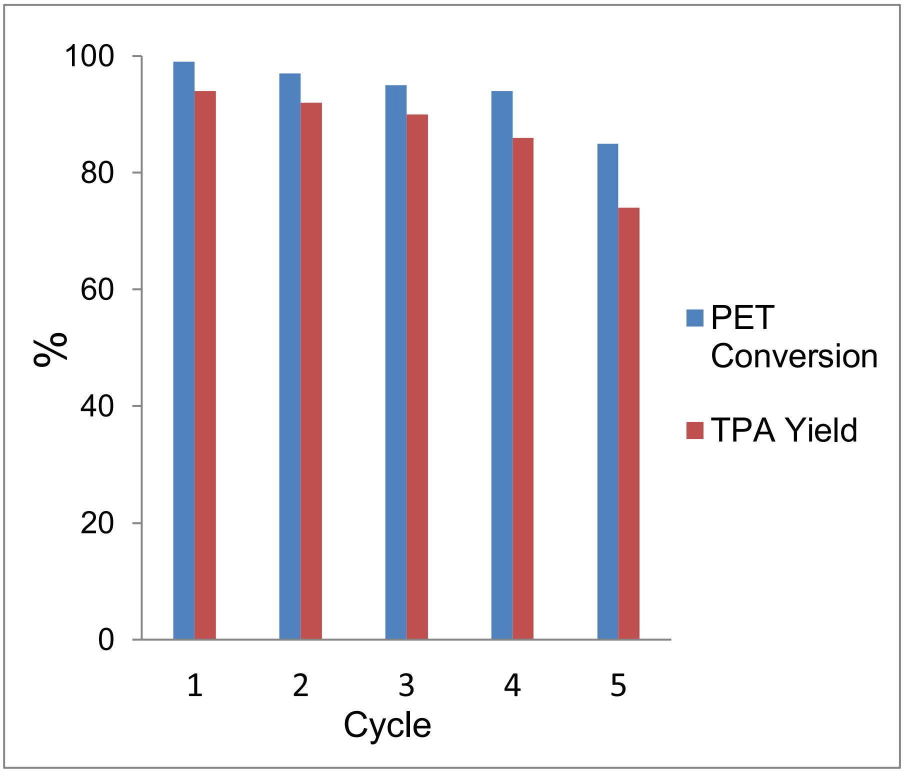

Fig. 3. *poly*(ethylene terephthalate) (PET) conversions and terephthalic acid (TPA) yields during the reuse of 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) catalyst in five catalytic cycles. All reactions were carried out using 0.500 g of PET powder (18 mesh), 5.00 mL of aqueous catalyst solution in 25 mL stainless steel solvothermal reaction kettles with Teflon inner sleeves, at 210 °C for 24 h.

The IR spectra and TGA profiles of isolated and authentic samples of terephthalic acid are included in the supplementary material as **S1** and **S2**.

2.3. Recovery and reuse of 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) catalyst in depolymerization of poly(ethylene terephthalate)

Recovery and reuse of 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) catalyst in polymerization of PET was tested for five catalytic cycles. The aqueous supernatant containing catalyst was separated from TPA by decanting the liquid after centrifugation, concentrated to 5.00 mL and reused for hydrolysis of PET as described in the general procedure 2.2. No attempts were made to separate ethylene glycol product from BAIL catalyst and the aqueous supernatant was recovered and reused for five successive catalytic cycles. The PET conversions and TPA yields in reusing 1.0 M aq. BAIL-1 catalyst under 210 °C, 24 h. reaction conditions in five catalytic cycles are shown in Fig. 3.

3. Results and discussion

3.1. BAIL Catalyzed depolymerization of poly(ethylene terephthalate) in water

1-(3-Propylsulfonic)-3-methylimidazolium chloride (BAIL-1), 1-(4-butylsulfonic)-3-methylimidazolium chloride (BAIL-2) catalysts were synthesized by condensation of 1-methylimidazole with C-3 and C-4 sultones, followed by acidification with HCl in 92 and 94 % yields respectively, following the literature procedures as shown in Fig. 2. (Gui et al., 2004), (Yang et al., 2008). The PET depolymerization experiments were carried out using 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1), 1-(4-butylsulfonic)-3-methylimidazolium chloride (BAIL-2) as shown in Fig. 1. In addition, the catalytic activities of these BAIL catalysts were compared with that of sulfuric acid and the results of all PET depolymerizations are shown in Table 1. The initial experiments at 210 °C were carried out using 0.05 and 0.10 M solutions of BAIL-1; these experiments resulted relatively low PET conversions as well as low TPA yields of 33 and 48% respectively after 48 h. as shown in entries 1 and 2 in Table 1. The increase in reaction time to 168 h., at a lower temperature 180 °C resulted even lower TPA yield of 39% (Entry 3). Then an increase in catalyst concentration to 0.50 M, and maintaining the temperature at 210 °C for 24 h resulted significant improvements in PET conversion to 70% and TPA yield to 65% (Entry 4). The best PET depolymerizations were achieved by using 1.00 M aqueous 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) catalyst at 210 °C, where a practically complete decomposition of PET and a 94% TPA isolated yield could be achieved after the 24 h. reaction time as shown in entry 5. In the next set of entries 6 and 7 we have compared the activities of two Brönsted acidic ionic liquid catalysts BAIL-1 and 2, by repeating the experiments 4 and 5 with the use of BAIL-2 as the catalyst. These experiments indicated that 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) is a better catalyst than 1-(4-butylsulfonic)-3-methylimidazolium chloride (BAIL-2) in depolymerization of PET in water at 210 °C. Interestingly, similar differences in catalytic activities of BAIL-1 and 2 are known in application of these catalysts in depolymerization of cellulose to glucose as well (Amarasekara and Owereh, 2009), (Amarasekara and Wiredu, 2014).

In the last set of experiments we have compared the catalytic activities of Brönsted acidic ionic liquid catalysts with sulfuric acid under similar H^+ ion concentrations and reaction conditions. The experiment using a low sulfuric acid concentration of 0.05 M resulted a poor conversion and a low PET yield of 18% as shown in entry 8. In assessment of entries 4 and 9 with $[H^+] = 0.50$ M, BAIL-1 catalyst produced a 70% PET conversion and a 65% TPA yield; whereas the comparable H_2SO_4 catalyst produced only 56% PET conversion and 52% TPA yield. Similarly, In assessment of entries 5 and 10 with $[H^+] = 1.00$ M, BAIL-1 catalyst produced a 99% PET conversion and a 94% TPA yield; whereas the comparable H_2SO_4 as a catalyst experiment produced only a 65% PET conversion and 63% TPA yield. These comparison experiments clearly prove the superior catalytic activities of sulfonic acid group functionalized Brönsted acidic ionic liquids in comparison to sulfuric acid. Imidazolium type ionic liquids are well known for their abilities to form charge-transfer complexes and hydrogen bonding type interactions with substrates and solvents (Izgorodina and MacFarlane, 2011), (Gonfa et al., 2011). In addition, a recent density functional theory (DFT) study on glycolysis of *poly*(ethylene terephthalate) in imidazolium ionic liquids, has revealed the importance of ionic interactions of the anions as well (Ju et al., 2018).

3.2. *Reuse of BAIL catalyst in depolymerization of poly(ethylene terephthalate)*

We have studied the recyclability of the BAIL catalyst by reusing the catalyst in five cycles. The catalyst was recovered by separation of the solid TPA product by centrifugation. However, ethylene glycol product was not separated from the recovered catalyst in water solution and the catalyst with increasing amounts of ethylene glycol in the medium was used in successive reuse experiments. The PET conversions and TPA yields during the reuse of 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) catalyst in five catalytic cycles are shown in Fig. 3. The PET conversion and TPA yields showed small, gradual decreases up to four cycles, though a significant reduction in the catalytic activity is seen in the fifth cycle. This steady decrease in PET conversions and TPA yields due to declining catalytic activity may be due to accumulation of the ethylene glycol product in the reaction medium.

The patented acid hydrolysis methods of PET are known since 1976, and concentrated H_2SO_4, or HCl is used as the solvent and catalyst in these techniques (Brown Jr and O'brien, 1976), (Pusztaszeri, 1982), (Sharma et al., 1985). For example, 78–90% (14.4–16.6 M) concentrated H_2SO_4 is proposed by Pusztaszeri, Brown, O'Brien and Sharma in their patents, in order to avoid high pressures and temperatures in the reaction vessel. Nevertheless, the major drawback of the strong acid process is the cost in recycling of large amounts of concentrated H_2SO_4 and generation of large quantities of inorganic salts and aqueous wastes. Yoshioka also described an acid catalyzed process, where highest PET degradation of 91.3% was reached in 13 M nitric acid at 100 °C for 24 h. and the resulting ethylene glycol was simultaneously oxidized to oxalic acid in this method, producing another value added product (Yoshioka et al., 1998). Liu et al. have studied the acid catalyzed depolymerization of PET in 1-*n*-butyl-3-methylimidazolium chloride ([BMIM][Cl]) as solvent and acid-functionalized ionic liquid 1-(3-propylsulfonic)-3-methylimidazolium hydrogen sulfate ([HSPMIM][HSO_4]) as catalyst. Under the optimum conditions of m(PET): m(H_2O): m(BMIM][Cl]): m([HSO_3-PMIM][HSO_4]) = 3: 4: 6: 0.6, and reaction temperature, 170 °C for 4.5 h, they have reported a 100% PET conversion and a 88% TPA yield (Liu et al., 2009). In comparison to these reported examples of using concentrated acids as well as acidic ionic liquids in neutral ionic liquid - water mixture mediums, the new BAIL catalyzed aqueous medium method has certain advantages like, recyclability of the acid catalyst, and avoiding the use of highly corrosive concentrated acids. The enhanced catalytic activity observed in Brönsted acidic ionic liquids is most likely due to their ability to interact or bind with the *poly*(ethylene terephthalate) through charge transfer and dipolar type interactions. In addition, a parallel catalysis activity enhancement of acidic ionic liquids in comparison to sulfuric acid is known in the case of BAIL catalyzed depolymerizations of cellulose to glucose in water as well (A.S. Amarasekara and Wiredu, 2011).

4. Conclusion

In conclusion, we have shown that sulfonic acid group functionalized imidazolium type Brönsted acidic ionic liquids are better catalysts in comparison to sulfuric acid with similar acid concentration for depolymerization of *poly*(ethylene terephthalate) in hot compressed water. The highest activity was found when using 1.00 M aqueous 1-(3-propylsulfonic)-3-methylimidazolium chloride (BAIL-1) catalyst at 210 °C, where a practically complete decomposition of PET and a 94% TPA isolated yields could be achieved after 24 h. The enhanced catalytic activity of Brönsted acidic ionic liquids in comparison to sulfuric acid with similar acid strength is most likely due the charge-transfer complex formation and hydrogen bonding type interactions of acidic ionic liquid catalysts with *poly*(ethylene terephthalate). Furthermore, Brönsted acidic ionic liquid catalyst could be reused for up to four cycles without a significant loss in catalytic activity.

CRediT authorship contribution statement

Ananda S. Amarasekara: Conceptualization and writing original draft. **Jay A. Gonzalez**: Laboratory experiments. **Victor C. Nwankwo**: Laboratory experiments.

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.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jil.2022.100021.

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