

1. Materials and Methods

1.1 Chemicals

The solvent, FT-IR spectrum of which is shown in Figure S1, used was an ionic liquid (IL) prepared by mixing a tertiary trialkyl amine (TEHA: tris(2-ethylhexyl)amine) and a short-chain carboxylic acid (DMBA: 2,2-dimethylbutyric acid), both purchased from Sigma-Aldrich. The DMBA and TEHA were mixed in a 1:1 ratio at room temperature following the method described in [1]. Isopropanol (IPA) (98%, industrial grade, Romil Ltd, Cambridge, UK) was used as an antisolvent. Virgin isotactic polypropylene (PP) was purchased from Sigma-Aldrich (UK).

Table S1. The carpet waste material was pre-consumer off-cuts provided by a UK carpet manufacturer. This was mechanically processed at Rotajet Systems Ltd., Wakefield, UK, through a sink & float process. The carpet off-cuts were fed into a unit equipped with a blade arrangement used to cut the carpet into small pieces. This was followed by a sink & float process to separate the PP fibres from other fractions, such as other polymers, additives and dirt, depending on their densities, facilitating the subsequent dissolution process (**Figure S2**).

Table S1. The manufacturer's datasheet provides the properties of virgin isotactic polypropylene.

Property	Value
Density (kg/m ³) at 25 °C	0.9
Molecular weight	
number averaged (M _n) (g.mol ⁻¹)	~67,000
weight averaged (M _w) (g.mol ⁻¹)	~250,000
Melt index (g/10 min) at 230 °C and 2.16 kg load	12
Melting point (°C)	160-165
CAS number	9003-07-0

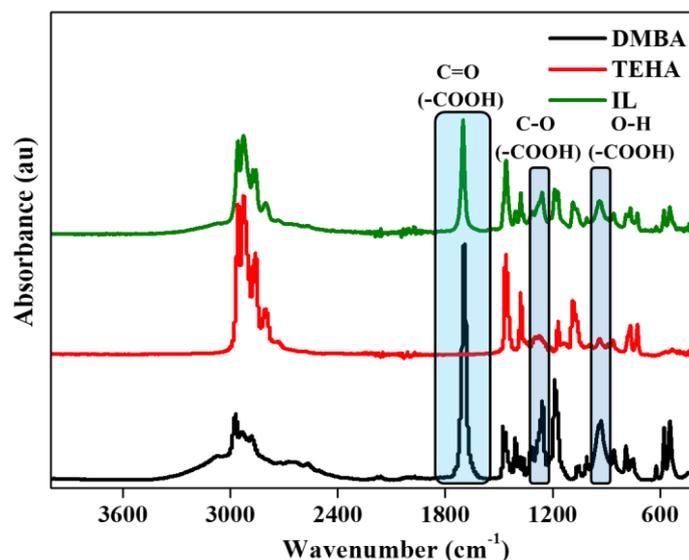


Figure S1. FTIR spectra of the IL used and its precursors.

FTIR characterisation (Fig S1) of the IL and precursors matched the typical spectra found as references for such substances. The strong peak at 1695.6 cm^{-1} is characteristic for C=O group of the carboxyl of the DMBA. It also appears at 1700 cm^{-1} in the IL. Once DMBA is mixed with TEHA, the C=O band signature is expected to shift to a higher wavenumber due to the formation of the ammonium salt between the DMBA and TEHA. The C-O and O-H vibrations characteristic of the -COOH functional group appear in both DMBA and IL samples at 1259 cm^{-1} , 920.2 cm^{-1} , 1279.7 cm^{-1} and 938 cm^{-1} , respectively. In this case, the shift to higher wavenumbers can be observed due to the interaction between the amine and acid. The presence of the O-H signal in the FTIR spectra of DMBA and IL confirms the presence of the -COOH.

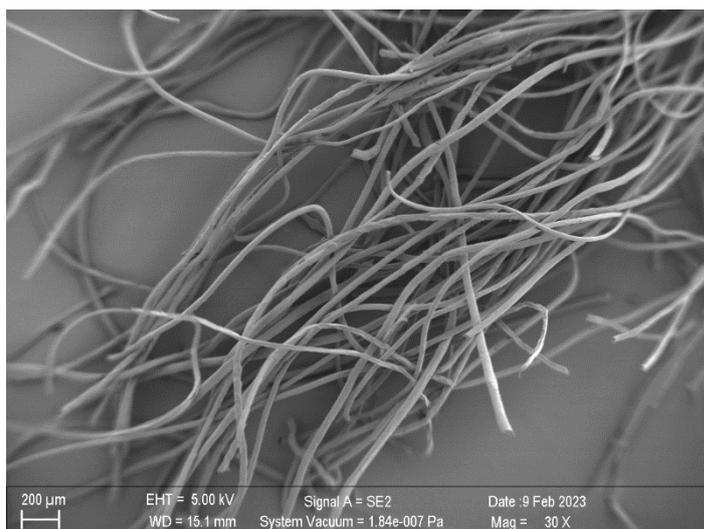


Figure S2. Scanning Electron Microscope (SEM) image of the polypropylene (PP) rich nonwoven carpet yarn.



Figure S3: Images of (left) nonwoven carpet yarn prior to dissolution and (right) recycled PP.

1.2 Preparation of solutions

The solubility of virgin PP in IL at three different temperatures, 140, 150 and 160 °C, aimed at determining an appropriate experimental dissolution temperature. This was conducted by pouring a volume of IL into a three-neck round bottom flask with a stirring bar and coupled to a reflux condenser, a thermometer and a glass needle through which gas nitrogen was circulated. The flask was flushed with nitrogen for about three minutes and then kept flowing during the experiment to maintain an inert atmosphere. Pellets of virgin PP were treated with liquid nitrogen and pulverised using a commercial grinder. Portions of the powder were added timely and spaced into the hot IL at the working temperature until the mixture reached about 13% wt. (g of PP/mL of IL). The solution was mixed at 900 rpm and maintained at a constant temperature until complete dissolution was observed. It was found that 160 °C was the temperature for the fastest dissolution of virgin PP powder (**Figure S3**) and was selected for this experimental work.

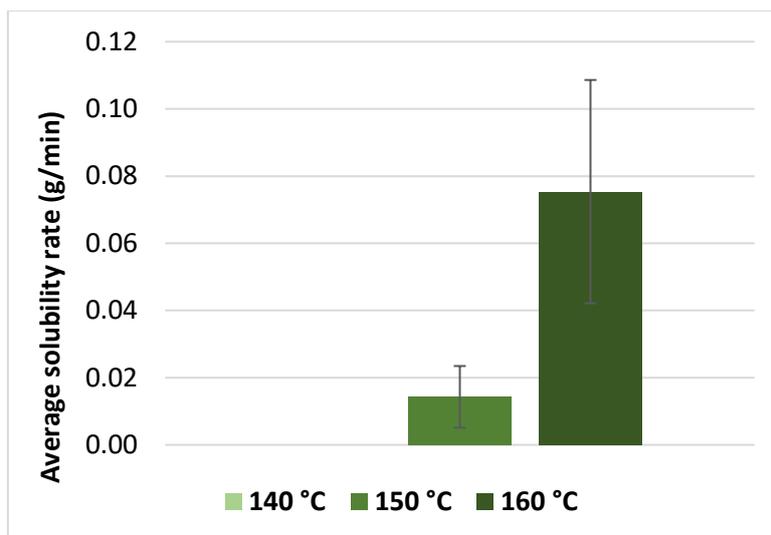


Figure S3. Solubility of virgin PP powder in IL at the temperatures used in the experiment. The standard deviation is shown as error bars. No dissolution was observed for the study at 140 °C after two hours. The magnitude of the standard deviation depicts the influence of the direct observation approach followed for the experiment.

1.3 Dissolution of virgin PP and PP-rich carpet waste

Pellets of virgin PP were sieved using a 4.75 mm mesh sieve to obtain an average particle size of ~3 mm. This particle size was selected to facilitate the sampling of the hot solution throughout the experiments. Preliminary work using pulverised virgin PP with a particle size similar than PP fibres exhibited a fast dissolution rate and challenging to follow. A volume of the IL was poured into a three-neck round bottom flask with the same setup described in the previous section. The solvent was continuously stirred and heated up to 160 °C. Portions of virgin PP and PP-rich carpet fibres were weighed and added to the solvent IL at 160 °C in proportion to obtain a concentration of 10% w/v. (g of virgin PP/mL of IL) and 12% w/v. (g of PP-rich carpet fibre/mL of IL). The previous solubility experiments conducted within this study demonstrated that working at higher concentrations makes the solution difficult to handle due to the high viscosity and interferes with the kinetics assessment.

1.4 Dissolution sampling for kinetics assessment

Several aliquots of the solution at 160 °C were manually taken timely spaced using a hot glass pipette and added into a volume of cold antisolvent, where the PP precipitated immediately. The precipitated polymer was then filtrated from the liquid mixture of IL and IPA using a cellulose membrane filter with a 0.45 µm pore size and washed several times with IPA. The recovered PP was dried overnight in an oven under full vacuum (1000 mbar) and weighed. The solvents were separated using a rotavapor under reduced pressure, and the volume of recovered IL was measured. The percentage of the recovered PP was calculated gravimetrically

considering the amount of polymer precipitated in the volume of IL in the individual sample using the following **equation (eq.S1)**:

$$\text{Recycled PP (RPP, \%)} = \frac{\text{Mass of reclaimed polypropylene (g)}}{\text{Volume of recovered ISOPREP liquid (mL)}} \times 100 \quad (\text{eq.S1})$$

1.5 Characterisation of virgin and reclaimed PP

Fourier-transformed infrared with attenuated total reflectance (FTIR-ATR) was performed using a Thermo Fisher Nicolet iS50 FTIR spectrometer with a built-in mid and far infrared capable diamond ATR in the wavenumber range between 4000 and 400 cm^{-1} to collect and compare the absorption spectra of virgin PP, carpet waste material (PP-rich fraction) and recycled PP. Pellets of virgin PP were treated with liquid nitrogen and pulverised using a commercial grinder to facilitate the measurement by the ATR sensor. For each spectrum, 16 single measurements were recorded and then averaged using the Omnic Spectra software.

High-Temperature Gel Permeation Chromatography (HT-GPC) tests were performed using an Agilent Infinity II High-Temperature GPC instrument equipped with differential refractive index (DRI), viscometry (VS) and dual angle light scatter (LS 90 + 15) detectors. The system was equipped with 2 x PLgel Olexis columns (300 x 7.5 mm) and a 10 μm guard column. The mobile phase was 1,2,4-trichlorobenzene (TCB) with 250 ppm butylated hydroxytoluene (BHT) additive. Samples were run at 1 mL/min at 160 $^{\circ}\text{C}$. Polystyrene standards (Agilent EasiVials) were used to create a third-order calibration. Analyte samples were dissolved at 140 $^{\circ}\text{C}$ and hot-filtered through a stainless-steel frit with 10 μm pore size before injection. Respectively, experimental molar mass (M_n ,SEC) and dispersity (\mathcal{D}) (or polydispersity index (PDI)) values of synthesised polymers were determined by conventional calibration using Agilent GPC/SEC software. Origin software was used for data processing.

Thermogravimetric analysis (TGA) was conducted using a Mettler Toledo TGA/DSC 3+ to investigate the weight loss behaviour of the virgin polymer, PP-rich waste material and reclaimed PP. A portion of the specimen was placed into a 70 μL alumina crucible and heated from 25 $^{\circ}\text{C}$ to 600 $^{\circ}\text{C}$ at a heating rate of 10 $^{\circ}\text{C}/\text{min}$ under a flow of nitrogen (50 mL/min). The data on weight loss and temperature were collected and processed using the STARe and Origin software, respectively.

Differential Scanning Calorimetry (DSC) tests were performed using a TA Instruments DSC 2500 equipped with liquid nitrogen to determine the crystallisation and melting temperatures of the original and recovered PP specimens. The method was run with nitrogen gas at 20.0 mL/min, and the samples were made of approximately 5 mg of the specimen in hermetic 40 μL aluminium pans, with a pinhole in the top to allow for gas evolution. The pans were heated and cooled between -30 and 280 $^{\circ}\text{C}$ at a rate of 10 $^{\circ}\text{C}/\text{min}$ in an atmosphere of nitrogen, and the data was processed through the TRIOS and Origin software.

2. References

- 1 Eur. Union, *EP2322497*, 2005.

