



Research article

Recycling of enamelled copper wire from end-of-life electric motor via room temperature methanolysis

Samuel D. Widijatmoko^{a,*}, Zhehao (John) Cui^b, Hassan Agalit^a, Yongliang Li^a, Gary A. Leeke^a

^a School of Chemical Engineering, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom

^b Grantham Research Institute on Climate Change and the Environment, London School of Economics (LSE), 32 Lincoln's Inn Fields, London, WC2A 3PH, United Kingdom

ARTICLE INFO

Keywords:

Enamel
Recycling
Copper
Wire
Polymer
Waste
Solvolytic

ABSTRACT

Polyester enamelled copper wire plays an important role in the manufacturing of electric motors. In line with the electrification of transport, the demand for electric motors and the future waste generated from their end-of-life cannot be ignored. The waste from the polyester enamelled copper wire is expected to increase steadily. Methods proposed by researchers are mainly focused on thermal treatment to either pyrolyse or burn off the polyester enamel. However, thermal treatments fail to consider the potential risk of air pollution and to recover the polyester enamel.

In this manuscript, we propose two-stage processes comprised of methanol washing and room temperature methanolysis with dichloromethane as co-solvent and K_2CO_3 as catalyst to delaminate multilayered type enamelled copper wire. The methanol washing recovers polyvinyl butyral as it is, via dissolution. Whereas the methanolysis products are dimethyl terephthalate (DMT) and dimethyl isophthalate (DMI) which are precursors to the polyester and could be used to make new polyester. At room temperature, the parameters of solid to liquid, DCM to methanol, and K_2CO_3 to Cu_{wire} ratio, of 500 g/L, 1.00 mol/mol, and 0.10 wt%, respectively, allow complete removal of polyester enamel in 24 h. The methanolysis parameters described manage to give a modest DMT and DMI yield of 86.0% and 92.2%, respectively. The reaction time can be sped up by increasing the temperature by 10 °C, leading to complete depolymerisation in 4 h. Compared to thermal treatment, the proposed method requires 80.7% lower energy with the products contained within the solution.

1. Introduction

The research of end-of-life EVs can be classified into three main broad topics of battery, traction motor, and power electronics (Elwert et al., 2015). Researchers are mainly targeting batteries as this is where the main recycling incentives lie (Nurdiawati and Agrawal, 2022). In contrast, only a few studies are focussing on electric motors and power electronics (Elwert et al., 2015). Hence, there is the need for more research aimed at electric motors and power electronics to enable close-loop EV recycling.

Tight windings in electric vehicles' motors, transformers, and any other types of electromagnetic equipment require enamelled copper wires as their main component. The enamel is a multi-layer of either a single type or a combination of polymeric insulators such as polyester (Liu et al., 2020), polyester imide (Petitgas et al., 2011), polyurethane (Ueda, 1989), polyimide (Petitgas et al., 2011) and poly(amide-imides) (Murray, 2008), with a typical thickness of 2 µm for each layer (Petitgas et al., 2011) up to a total thickness of 20 µm (Haque et al., 2014).

Having excellent processability, adhesive properties, good flexibility, and a simple process of synthesis, polyester enamels have been the most widely used for over 50 years. In 2008, the production of polyester enamelled copper wire was reported to be at least 50,000 tonnes a year or equivalent to 15% of the total enamelled wire in the world (Bhanu, 2008). Currently, it is estimated that the end-of-life polyester enamelled copper wire has reached millions of tonnes (Ma et al., 2023). Findings by Ma et al. (2023) showed that waste enamelled copper wire may contain 3.0 wt% polyester which translates to at least 30,000 tonnes of polyester needing to be recycled for every million tonne of waste enamelled copper wire. Recycling 30,000 tonnes of polyester from waste enamelled copper wire, taking into account the embodied carbon of manufacturing polyester which is 32 kg CO₂ eq/kg (Kilgore, 2023), saves at least 960,000 tonnes of CO₂ eq. Due to the enamel being closely attached to the copper wire and difficult to remove, recent studies to recycle waste enamelled copper wire only focus on thermal treatment. The thermal treatment proposed involves high temperature or pyrolysis under an inert atmosphere with temperatures ranging from 500 °C to 900 °C (Li et al., 2023; Liu et al., 2020) or directly applying

* Corresponding author.

E-mail addresses: s.d.widijatmoko@bham.ac.uk (S.D. Widijatmoko), g.a.leeke@bham.ac.uk (G.A. Leeke).

a voltage to heat the copper wire above the decomposition temperature of the enamel (Mominul Haque et al., 2012). However, both methods did not consider the recycling of the polyester enamel as well as the potential release of harmful gases into the environment such as CO, CO₂, and various hydrocarbons by-products.

Chemical recycling of polymer involves the depolymerisation of polymer chains. The original monomers or oligomers can be re-claimed, creating a secondary virgin quality raw material (Barnard et al., 2021). Solvolysis is a chemical recycling pathway that depolymerises polymers using solvents such as water (hydrolysis), alcohols (alcoholysis), amines (aminolysis), ammonia (ammonolysis), and glycol (glycolysis) (Shirazimoghaddam et al., 2023). Except for glycolysis, the solvolysis of polyester (i.e., polyethylene terephthalate/PET) produces ethylene glycol (EG) as a by-product (Jiang et al., 2022). The EG can be separated from the monomers by distillation (Ishihara et al., 2004).

Alcoholysis using methanol (methanolysis) is a process that is typically carried out at high pressure and temperature to decompose PET into dimethyl terephthalate (DMT) and EG, which are the raw materials necessary for the production of this polymer (Paszun and Szychaj, 1997). The commercial process is performed at temperatures between 180 °C to 280 °C and pressures between 2 MPa to 4 MPa (Scheirs, 1998). Due to the low solubility of DMT in water, it can be easily purified to an acceptable grade that is applicable as an alternative feedstock to produce PET (Pudack et al., 2020).

Methanolysis for PET recycling has been extensively studied. Some notable processes include; depolymerisation under supercritical methanol (<1 h, 260 °C – 270 °C, 9 MPa – 11 MPa) (Genta et al., 2005; Yang et al., 2002), microwave assisted methanolysis (0.5 h, 160 °C) with zinc acetate as the catalyst (Siddiqui et al., 2012), and the use of co-solvents such as halogenated solvents to swell PET with alkali decomposition, which enables methanolysis to be carried out at room temperature (Essaddam, 2020). Recently, Pham and Cho (2021) have studied the role of different co-solvent and alkali catalysts for room temperature methanolysis of PET. They report that the best co-solvent and catalyst for room temperature methanolysis are dichloromethane (DCM) and K₂CO₃, respectively.

In this manuscript, we applied a room temperature methanolysis with DCM as co-solvent and K₂CO₃ catalyst for the facile delamination of polyester enamelled spent copper wire. Compared to thermal treatment, the proposed process requires lower energy expenditure and does not produce harmful gas products.

2. Materials and methods

2.1. Copper wire sample and characterisation

Waste 0.27 mm enamelled copper wires were obtained from EMR Metal Recycling Birmingham. The collected enamelled copper wires were uncoiled manually and chopped into 1 cm pieces.

The chopped enamelled copper wires were first analysed for purity. Sample copper wire is weighed to 4 decimal places and digested using a mixture of 3 parts 32% HCl and 1 part of 68% HNO₃. The solution was diluted and analysed using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS, PerkinElmer-NexION 300x) with helium kinetic energy discrimination (KED) mode. Multi-elements calibration solutions were made from a certified reference material from Merck by serial dilution 1 ppb – 5000 ppb; ICP multi-element standard solution IV (Lot No. HC15457555). Internal standardisation was carried out using Rhodium-103 and added automatically by the system to correct the instrument for drift and physical interference. Calibration was carried out for the following elements: Li-7, B-11, Ag-107, In-115, Ba-138, Tl-205, Pb-208, Bi-209, Na-23, Mg-24, Al-27, K-39, Ca-43, Cr-52, Mn-55, Fe-56, Co-59, Ni-60, Cu-63, Zn-66, Ga-69, Sr-88, Cd-111.

Surface characterisation was done using Fourier Transform Infrared Spectroscopy-Attenuated Total Internal Reflection (FTIR-ATR, Thermo Scientific Nicolet Summit X). All measurements were carried out with

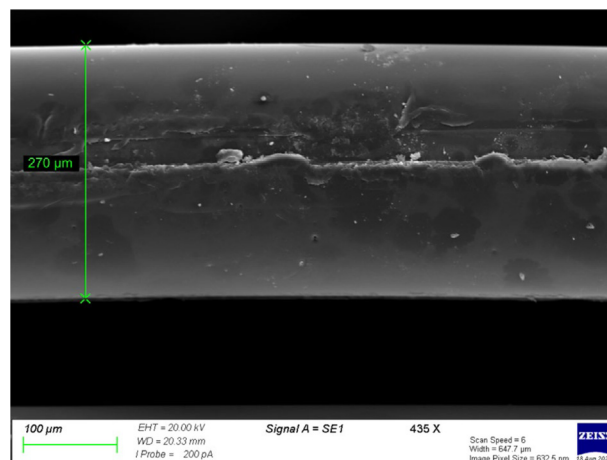


Fig. 1. SEM image of as received waste enamelled copper wire.

24 scans and a resolution of 4 cm⁻¹. The spectra collected are compared to the Sigma-Aldrich Library of ATR-IR Spectra (Wiley).

Morphological analysis was carried out using a Scanning Electron Microscopy (SEM, Zeiss EVO 10). All samples were mounted onto aluminium studs using carbon tape and the surface was made conductive using a gold sputtering machine (Quorum Q 150R ES) with the thickness programmed at 10 nm. The morphology of intact enamelled copper wire is presented in Fig. 1.

2.2. Methanol wash procedure and analysis

A certain amount of enamelled copper wires was mixed with a certain amount of methanol and left standing for a certain amount of time. The washed copper wires were separated from the solution via filtration and dried in a fume hood. Moreover, during washing, the liquid is occasionally sampled and analysed using an Ultraviolet-Visible spectrophotometer (UV-Vis, Agilent Cary 60) with a 1 cm quartz cuvette (Hellma) and scanned from 200 nm to 400 nm.

2.3. Methanolysis procedure and analysis

In a round bottom flask with a PTFE magnetic stirring bar, a certain amount of enamelled copper wire was mixed with a certain amount of anhydrous K₂CO₃ (ThermoFisher Scientific), methanol (HPLC grade-Fisher Chemicals), and DCM (Reagent Grade-Fisher Chemicals). The round bottom flask was sealed with Suba-Seal[®] and put onto a hotplate stirrer set at 25 °C and 300 RPM (Asynt – ADS-HP-NT with DrySyn[®] MULTI) for a certain amount of time. Moreover, the hot plate stirred was connected to a power meter (RS PRO – No. 1785370) to measure the total electricity consumed. The mixture then was filtered to recover the copper wires and the liquid product.

The liquid product then was analysed using a Gas Chromatography–Mass Spectrometer (GC–MS, ThermoFisher Trace 1600 – ISQ7610) with an Rtx-35sil (Restek) column. The mass spectrometer helps confirm the presence of DMT and DMI after methanolysis by comparing the mass spectrum fragmentation to that of the National Institute of Standards and Technology (NIST) mass spectral libraries.

The copper wires were washed with deionised water, to remove the K₂CO₃, followed by methanol, and air-dried in a fume hood. For screening purposes, the bulk density was assessed by using a helium gas pycnometer (Micromeritics AccuPyc II 1340). Due to the lower density of the enamel compared to the copper core, the bulk density of the copper wire sample would increase as the enamel is removed.

Dimethyl terephthalate (>99%, VWR), dimethyl isophthalate (>98%, Alfa Aesar), and dodecane (>99%, Sigma Aldrich) were purchased to calibrate the GC–MS for quantitative analysis. The quantification is based on the relative response factor of dodecane internal

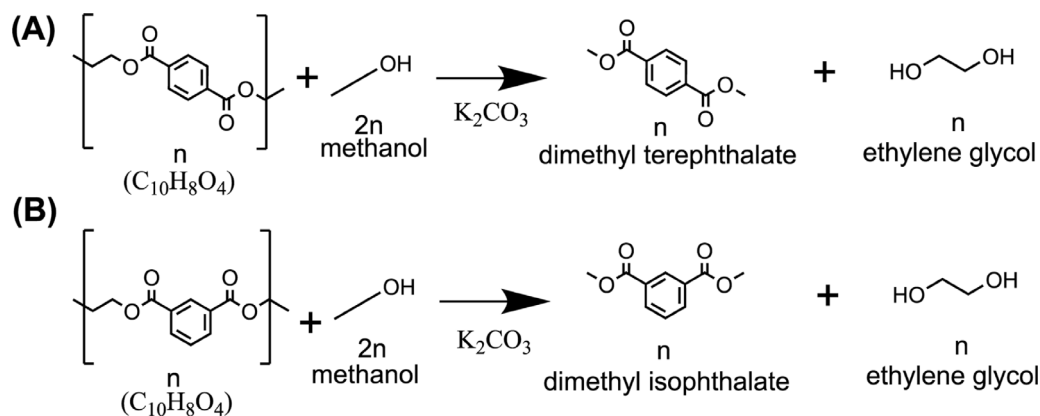


Fig. 2. Reaction step for polyester depolymerisation via methanolysis of (A) terephthalate repeating unit and (B) isophthalate repeating unit.

standard, to that of DMT and DMI. The ethylene glycol and oligomers by-product were not measured by this technique.

$$\text{Response Factor (RF)} = A/C \quad (1)$$

where C is the concentration of a specific compound and A is the resulting peak area obtained from the GC-MS chromatogram.

A mixture of known amounts of dodecane, dimethyl terephthalate, and dimethyl isophthalate was measured using GC-MS. The response factor of each different compound is calculated using Eq. (1). The relative response factor between different components can then be calculated using Eq. (2).

$$\text{Relative Response Factor (RRF)} = \text{RF}_A/\text{RF}_B \quad (2)$$

The concentration calculation for unknown analyte (C_A), given a known reference concentration (C_B) is described in Eq. (3).

$$C_A = A_A/A_B \times 1/\text{RRF} \times C_B \quad (3)$$

Thus, the concentration of DMT and DMI from an unknown sample can be calculated by mixing a known amount of unknown sample with a known amount of dodecane internal standard stock solution, which in this study is 1 mL unknown sample with 1 mL 2.5 g/L dodecane stock solution. The concentration of DMT and DMI from an unknown sample is the measured concentration times dilution factor that is equal to 2.

The methanolysis reaction step and mol balance for polyester containing terephthalate and isophthalate repeating unit are presented in Fig. 2. The measured concentration of DMT and DMI in the liquid is then used to calculate yield as shown in Eq. (4).

$$Y_A = N_A/N_o \times 100 \quad (4)$$

where N_A is the number of moles of DMT or DMI and N_o is the number of moles of repeating units; with the molar ratio of terephthalate and isophthalate repeating unit was calculated based on the molar concentration ratio of the DMT and DMI in the product.

2.4. Nuclear magnetic resonance spectroscopy

The solutions obtained after methanolysis procedure were transferred to a 250 mL round bottom flask and subjected to a rotary evaporator (Stuart-RE400/MS) maintained at 40 °C to remove the DCM and methanol. After most of the solvents had been removed, the sample was left to cool down to room temperature and 1 mL of deuterated chloroform (99.8%, Cambridge Isotope Laboratories, Inc.) was added and gently rinse to dissolve the product. The solutions were then collected and transferred to a Nuclear Magnetic Resonance (NMR) tube (Norell®-NOR508UP7-5EA) and analysed using a Bruker-400 MHz NMR spectrometer. The NMR was used to confirm that the reaction is complete (see supplementary information) and confirm that the products formed are indeed DMT, DMI and EG.

2.5. Thermogravimetric analysis

Calcinations were carried out using a simultaneous TG-DTA/DSC apparatus (STA, NETZSCH - STA 449 F3 Jupiter) to determine the weight amount of enamel laminating the copper wire from different treatment stages. The calcination process was carried out from 25 °C to 900 °C with a heating rate, nitrogen gas flow rate, and final holding time of 10 °C/min, 50 mL/min, and 1 h, respectively.

The nitrogen atmosphere was selected to avoid copper oxidation with the implication of incomplete combustion of the polymer. The previously reported study of pyrolysis of polyester and polyvinyl butyral (PVB) polymer under a nitrogen atmosphere achieved completion at a temperature above 600 °C that resulted in 97.6 wt% (Liu et al., 2020) and 95.0 wt% (Tang et al., 2014) mass loss, respectively. Hence, the error due to the incomplete combustion was assumed to be negligible. The results from the thermogravimetric analysis from this study are presented in Fig. 3.

2.6. Energy consumption calculation

By using a simple energy consumption model, the overall energy consumed per kilogram of waste enamelled copper wire treated between the proposed and the previously reported thermal treatment were compared. The recent UK average commercial electricity tariff of 25p/kWh (AquaSwitch, 2023) and national grid average carbon emission factor of 0.21 kg CO₂eq/kWh (ITPENERGISED, 2023) were used in the model to calculate the electricity cost and resulting embodied carbon for both methods.

Fig. 4 present flow diagrams of the proposed process (this research) and the pyrolysis process proposed by Liu et al. (2020). The main unit operations considered for energy consumption of the proposed process are a mixer for washing and two rotary evaporators (rotavap) for solvent recycling by distillation. The proposed process comprises two stages of methanol washing followed by methanolysis and summarised in Fig. 4(A). In the first stage, the waste enamelled copper wire was subjected to methanol washing, stream S 1, to remove the outermost coating. The resulting output is partially cleaned copper (S 2) and methanol containing dissolved polymer (S 4). The methanol is recycled by distillation using a rotavap and re-used (S8-R) for more washing while the dissolved polymer can be further treated (S 7). In the second stage, the partially cleaned copper (S 2) is subjected to methanolysis to break down the polyester enamel to its monomer. After filtration, the resulting output is clean copper with a small amount of K₂CO₃ (S 3) and a mixture containing monomers dissolved in DCM and methanol (S 6). The methanol is then recycled by distillation using a rotavap and re-used (S10-R) for more reaction. It is important to point out that some methanol is being consumed for methanolysis and the K₂CO₃ catalyst is not recovered. Hence, methanol and K₂CO₃ top-up is required (S 11)

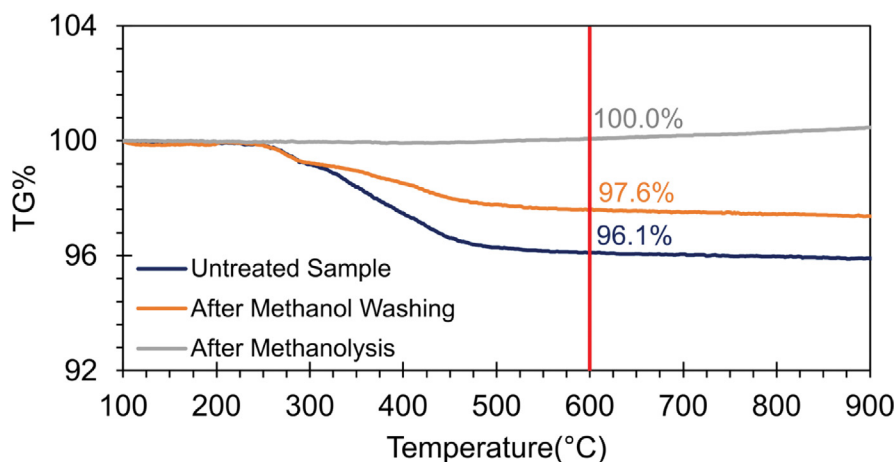


Fig. 3. TG curves of untreated copper wire, after methanol washing and after methanolysis.

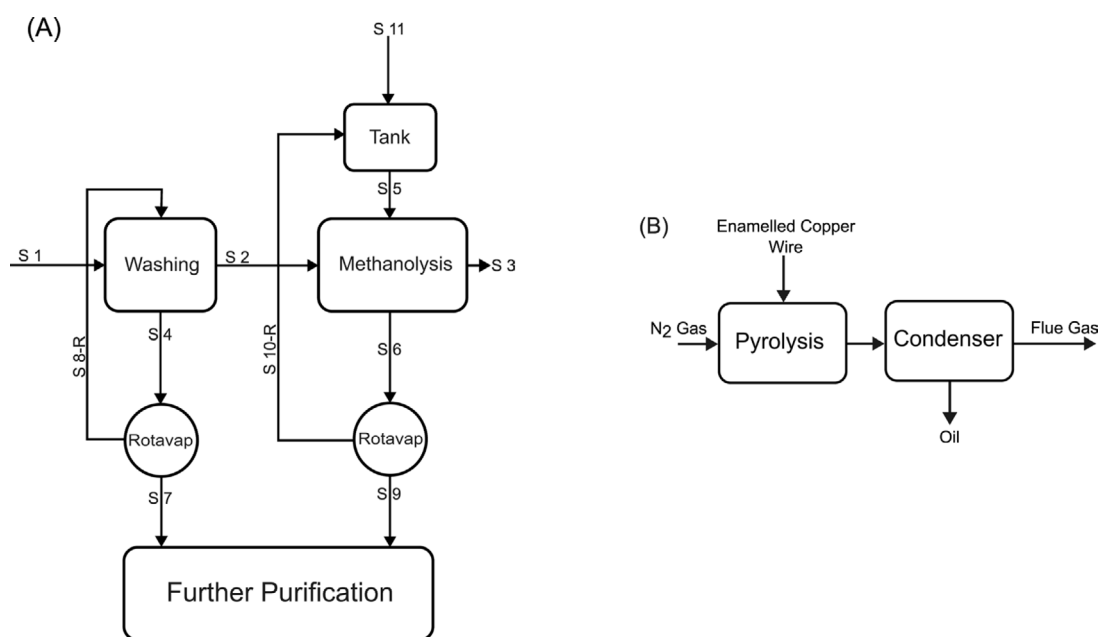


Fig. 4. Flow diagram of (A) the proposed process (this research) and (B) pyrolysis (Liu et al., 2020).

and mixed with the recycled solvent before further methanolysis (S 5). Whereas the main unit operation for pyrolysis considered for energy consumption is the electric furnace. From Fig. 4(B), the pyrolysis process involves the heating of enamelled copper wire at 600 °C under a nitrogen atmosphere. The resulting gas is passed through a passive condenser to collect the pyrolysis oil.

3. Results and discussion

3.1. Enamel characterisation and removal

The copper wire was analysed for coating type using ATR-FTIR and the spectrum for the as-received sample is presented in Fig. 5(A). The spectrum search via the Sigma-Aldrich Library of ATR-IR Spectra suggests that the polymer is poly (vinyl butyral-co-vinyl alcohol-co-vinyl acetate). From Fig. 5(A), the wide peak at wavenumbers 3405 cm^{-1} is the O-H stretching (Yang et al., 2013). The two peaks at 2922 cm^{-1} and 2871 cm^{-1} can be assigned to the stretching vibration of C-H (Luan et al., 2023). The peak at 1732 cm^{-1} is the G=O stretching. The peaks at 1433 cm^{-1} , 1378 cm^{-1} , and 1342 cm^{-1} are the C-H bending vibration (Qin and Cheng, 2016). The peaks at 1130 cm^{-1} and

1054 cm^{-1} are the C-O-C-O-C stretching vibrations of the cyclic acetal group (Qin and Cheng, 2016). The peaks at 990 cm^{-1} and 1240 cm^{-1} are the C-O-C stretching vibration of the acetate group (Qin and Cheng, 2016). From the band assignment, the spectrum obtained by ATR-FTIR is the PVB polymer.

It has been reported that PVB polymer is soluble in methanol (Samide et al., 2020). In principle, a simple methanol washing can remove the enamel. To confirm this, 2 g of the enamelled copper wires were immersed in 4 mL of methanol and left standing for 2 h. The copper wire was then collected via filtration, air dried, and analysed using ATR-FTIR.

The ATR-FTIR spectrum for the methanol washed copper wire sample is presented in Fig. 5(B). The ATR-FTIR spectrum after washing is different than that of the spectrum before washing. This indicates the presence of another polymer, and the enamel is of a multi-layer type with PVB polymer as its outermost layer. The spectrum search reveals that the polymer is a polyester type and is a combination of PET Fig. 5(B) and poly (diallyl isophthalate) Fig. 5(C).

From Fig. 5(B), the peak at wavenumbers 1705 cm^{-1} is the G=O stretching (Miyamae et al., 2001). The peak at wavenumbers 1215 cm^{-1} is the aromatic ether C-O stretching (Donelli et al., 2010). The

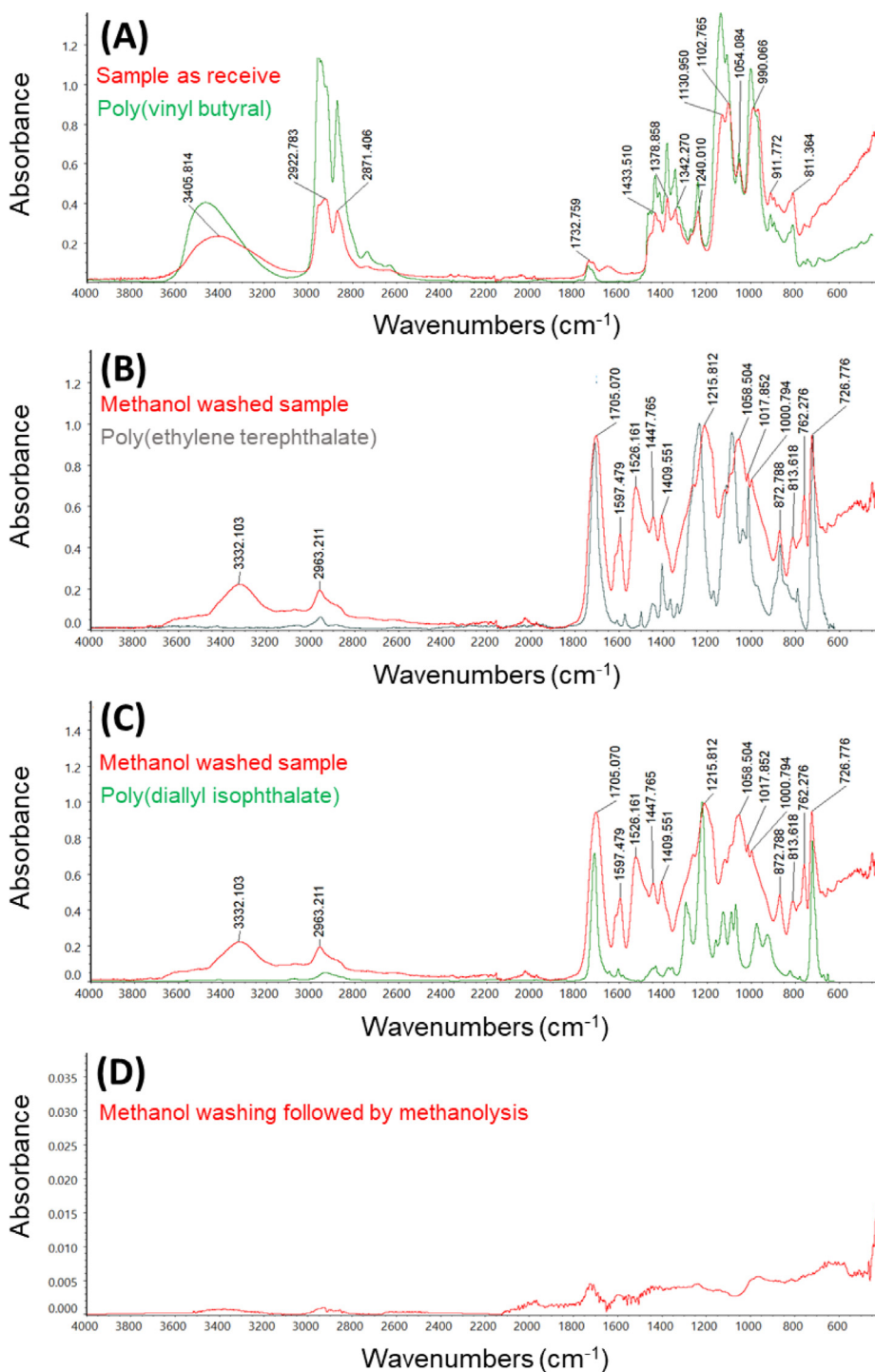


Fig. 5. ATR-FTIR spectrum of the waste enamelled copper wire compared to the Sigma-Aldrich Library of ATR-IR Spectra (Wiley) for (A) as received, (B) and (C) after methanol washing, and (D) after methanolysis.

peaks at wavenumbers 872 cm⁻¹ and 726 cm⁻¹ are the aromatic out of plane bending vibration (Donelli et al., 2010). The peaks at 1705 cm⁻¹ and 1215 cm⁻¹ indicate the ester functional group. While the peaks at 872 cm⁻¹ and 726 cm⁻¹ confirm the presence of an aromatic ring. The type of polymer is confirmed to be polyester type.

Furthermore, from Fig. 5(B), there are other peaks with significant absorbance that are not typical of polyester such as relatively strong absorption at 3332 cm⁻¹ and 1447 cm⁻¹ which are the stretching vibrations of O-H groups and alkane C-C stretch, respectively. It has previously been reported that degraded PET tends to have relatively higher absorbance in the region of 2700 cm⁻¹ to 3500 cm⁻¹ and

1400 cm⁻¹ to 1450 cm⁻¹ (Ioakeimidis et al., 2016). Hence, the ATR-FTIR spectrum also reveals that the polyester enamel has undergone severe degradation.

Knowing that the enamel is a polyester type, depolymerisation of polyester via room temperature methanolysis was proposed. Room temperature methanolysis requires DCM as a co-solvent and K₂CO₃ as a catalyst (Pham and Cho, 2021). 3.2917 g of washed enamelled copper wire is mixed with 6 mL of methanol, 5 mL of DCM, and 0.1215 g of K₂CO₃ catalyst or equivalent to S/L, DCM/Methanol, and K₂CO₃/Cu_{wire} ratio of 300 g/L, 0.53 mol/mol and 3.8 wt%. The mixture remained under stirring for 18 h at 25 °C. A copper wire was sampled from the

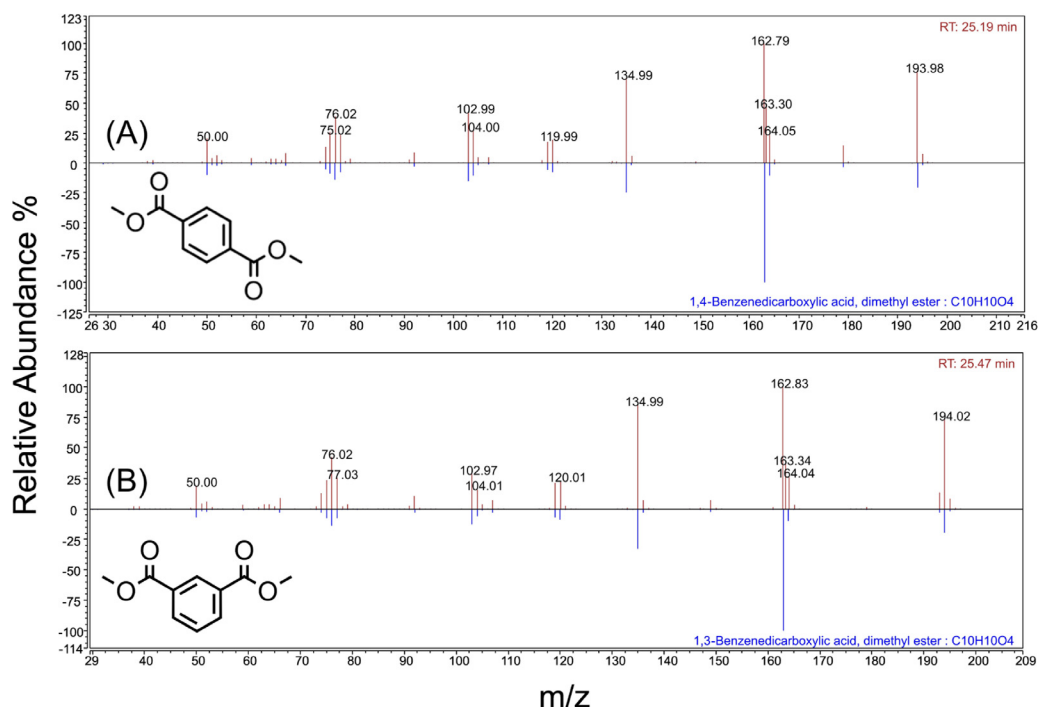


Fig. 6. Measured mass spectral (positive y-axis) versus NIST mass spectral libraries (negative y-axis) of (A) dimethyl terephthalate and (B) dimethyl isophthalate.

mixture, washed with methanol, air dried, and analysed using ATR-FTIR. The resulting spectrum is presented in Fig. 5(D) and it was found that a substantial amount of the polyester enamel had been removed.

To determine the copper core size and coating thickness, the diameter of the copper wire samples from different treatments were measured using a micrometre screw gauge accurate to 1 μm . From the average of five measurements, it was found that the diameter of the as-received, methanol washed, and methanol washed followed by methanolysis samples were 270 μm , 260 μm , and 235 μm , respectively. The measurements obtained from the morphological analysis were also in agreement with the results from the micrometre screw gauge. Hence, the PVB and polyester enamels have different thicknesses of 5 μm and 12.5 μm , respectively.

To determine the ratio of the terephthalate and isophthalate repeating unit, the liquid from the methanolysis described above was sampled and analysed using a GC-MS. The mass spectrum fragmentation of the two most significant peaks is presented in Fig. 6(A) and (B) and compared against that of the NIST mass spectral libraries. The two main products from the procedure were found to be DMT and DMI and confirmed the hypothesis that the polymer is polyester. Moreover, by using the relative response factor of dodecane to DMT and DMI, it was found that the molar ratio between DMT and DMI is unity.

Having shown that room temperature methanol washing and methanolysis with DCM co-solvent are useful techniques to delaminate the PVB and polyester enamel. A two-stage process to recover PVB by dissolution and polyester as DMT and DMI by methanolysis are proposed and summarised in Fig. 7. The density of copper wires from different treatments was measured and compared. The copper wires that underwent methanol washing and methanolysis have a measured density of 8.94 g/cm^3 , which is close to that of pure copper (ca., 8.94 g/cm^3). The same sample was digested and analysed for metal concentration using an ICP-MS. The result suggests that the sample contains 99.5 wt% copper. These findings ascertain that the enamelled copper wire was made of high purity copper.

The copper wires from different treatments are also subject to pyrolysis in an STA to quantify the amount of enamel present. From the thermogravimetric analysis, it can be deduced that the methanolysis had completely removed the polyester enamel. Moreover, the weight

percentage ratio between PVB, polyester, and copper core can be estimated and calculated to be 1.5 wt%, 2.4 wt%, and 96.1 wt%, respectively.

3.2. Parameters affecting enamel removal

The first stage of the proposed process involves dissolving PVB in methanol referred to as methanol washing. Methanol is selected due to the good solubility of PVB in alcohol (Paul and Cotts, 1987) and has been reported to be as much as 10.0 wt% (Peer et al., 2014) to 38.0 wt% (Ali et al., 2022). Moreover, PVB in solution exhibits weak and broad absorption bands between 250 nm to 300 nm (Posavec et al., 2014). Thus, making UV-Vis a suitable technique to monitor the dissolution of PVB enamel into methanol.

The time taken to fully remove PVB was studied by measuring the absorbance of the liquid methanol from various washing times. 2 g of received enamelled copper wire was weighted to four decimal places, transferred to a scintillation vial followed by the addition of 2.5 mL methanol, and sealed. The amount of methanol selected is equivalent to a weight ratio of enamelled copper wire to methanol of one and a complete PVB enamel dissolution results in a concentration of 1.5 wt% PVB in methanol. The timer was started as soon as the methanol was added. After 1 min, using a plastic pipette transfer, the liquid was transferred to a quartz cuvette and the absorbance was measured using a spectrophotometer. The procedures were repeated for 5 min and 30 min washing time, and the results are presented in Fig. 8.

From Fig. 8, there is a minimum increase in absorbance from 1 min to 30 min washing time. This indicates that the washing process happens promptly which can be caused by the combination of a small amount of PVB coating and its excellent solubility in methanol. The increase in absorbance was thought to occur due to solvent loss from the longer washing time rather than more PVB polymer being dissolved. To confirm this, copper wire from 1 min washing time was sampled and analysed using an ATR-FTIR. It was found that the PVB coating had been removed revealing the spectrum of polyester enamel underneath after 1 min washing time.

Considering the low concentration of PVB only after single pass washing, it is important to consider re-using the methanol wash solution. Taking a PVB concentration limit of 10.0 wt%, the wash methanol

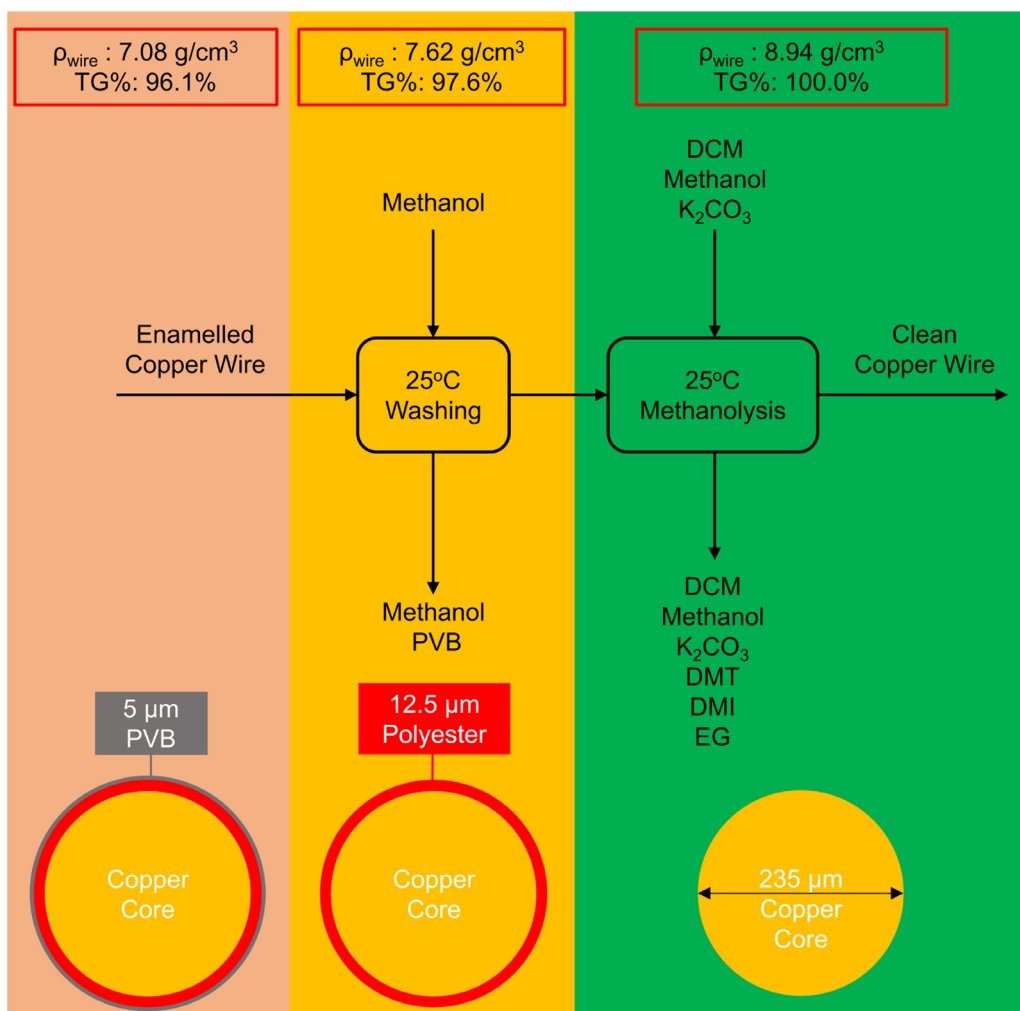


Fig. 7. Schematic diagram of the proposed process; illustrating enameled copper wire going through methanol washing and room temperature methanolysis.

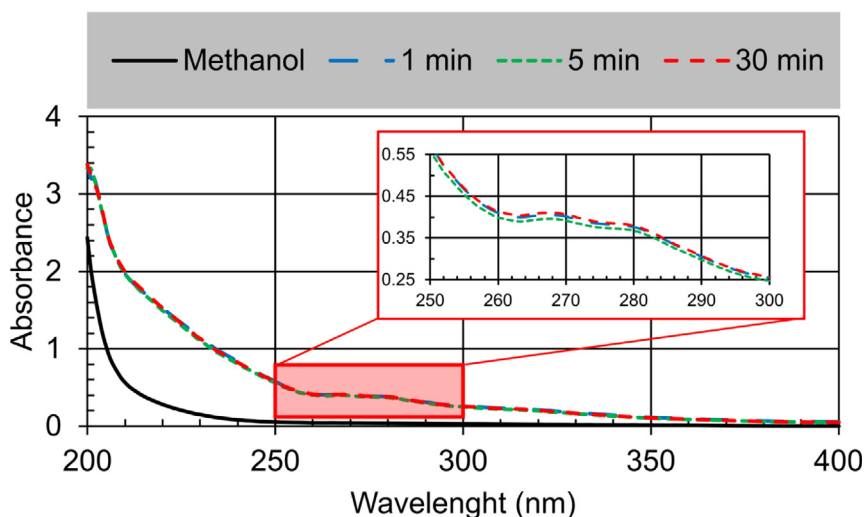


Fig. 8. UV-Vis spectrum of solutions from various washing times.

can be re-used for another 5 washing cycles. Taking a higher concentration limit caused the solution to become more viscous and recovering clean copper became more challenging.

The second stage of the process involves methanolysis at room temperature with DCM as co-solvent and K_2CO_3 as catalyst. Parameters

affecting enamel removal are studied by means of small-scale screening that considers solvolysis time between 0.25 h and 39 h, solid to liquid ratio (S/L) of 100 g/L to 500 g/L, catalyst to methanol washed copper wire ratio (K_2CO_3/Cu_{wire}) of 0.10 wt% to 7.5 wt%, and the molar ratio of DCM to methanol (DCM/Methanol) 0.05 mol/mol to 3.00 mol/mol

Table 1
Screening parameters and the measured density after treatment.

No.	Time (h)	S/L (g/L)	K ₂ CO ₃ /Cu _{wire} (wt%)	DCM/MeOH (mol/mol)	ρ_{measured} (g/cm ³)	$\rho_{\text{measured}}/\rho_{\text{Cu}}$ (%)
1	39	500	0.10	1.00	8.94	100
2	39	100	0.10	1.00	8.94	100
3	39	100	7.5	1.00	8.93	100
4	39	100	7.5	1.00	8.91	100
5	39	500	7.5	0.05	8.90	100
6	39	500	7.5	0.05	8.87	99
7	1	500	7.5	1.00	8.85	99
8	39	100	0.10	0.05	7.68	86
9	1	100	7.5	0.05	7.90	88
10	1	100	0.10	0.05	7.65	86
11	1	500	0.10	0.05	7.65	86
12	1	500	0.10	1.00	7.64	86
13	0.25	500	7.5	3.00	7.68	86
14	0.50	500	7.5	3.00	7.63	85

were carried out. The screening was done using an Assynt hot plate with inserts that can accommodate twelve 24.40 mm OD boiling tubes. Each boiling tube had a 5 mm × 12 mm stir bar and sealed with Suba-Seal®. The hotplate temperature and stirring were set at 25 °C and 300 RPM, respectively. After the treatment, the copper wires were recovered by filtration, washed with water followed by methanol, air dried, and analysed using a helium gas pycnometer. All samples used in the screening have been washed with methanol and free of PVB polymer. A complete enamel removal will result in a measured density increase from 7.62 g/cm³ to that of pure copper of 8.94 g/cm³.

The screening results are presented in Table 1 and reveal that only half (No. 1 to No. 7) of the screening parameters result in an almost complete removal of polyester enamel. From the parameters that allow almost complete removal of poly ester enamel, the parameters in Table 1 - No. 7 require the shortest duration with the expense of higher DCM/MeOH, S/L, and K₂CO₃/Cu_{wire} when compared to other screened parameters.

Too much catalyst is a waste of resources and minimising the use of DCM (Byrne et al., 2016) as a solvent is perceived as a good strategy to minimise the environmental impact related to its use. Further experiments were done to understand the effect of minimising the amount of K₂CO₃ catalyst and DCM towards the rate of polyester degradation. The rate of polyester degradation was analysed via off-line GC-MS analysis by monitoring the concentration of DMT and DMI over time. The parameters from Table 1 – No. 7 were used as a starting point (Run A) and the results are presented in Fig. 9(A).

The results presented in Fig. 9 were done in triplicate with each run using a basis of 50 g methanol washed enamelled copper wire. From Fig. 9(A), it can be observed that the yield did not further increase with increasing time beyond 1 h. This suggests that the reaction was completed in 1 h, resulting in 82.8% and 83.3% yield of DMT and DMI, respectively. The catalyst ratio was decreased from Run A of 7.5 wt% to 0.10 wt% (Run B) and the result is presented in Fig. 9(B). Comparing Run A to Run B there was a delayed reactivity as the yield of DMT and DMI at 1 h are 0.7% and 0.6% compared to 82.8% and 83.3%, respectively. In 24 h, the yields of DMT and DMI were 86.0% and 92.2%. In Run C, the DCM to methanol molar ratio was decreased from 1 to 0.2 mol/mol while keeping the other parameters the same as in Run B. Comparing Run C to Run B there was a further delayed reactivity as the yield of DMT and DMI at 3 h are 23.5% and 21.8% compared to 69.4% and 71.6%, respectively. In 24 h, the yield of DMT and DMI were 83.3% and 89.4%, respectively. Moreover, the copper wire density from Run A, Run B, and Run C were measured and found to be 8.94 g/cm³, 8.94 g/cm³, and 8.93 g/cm³, respectively. These findings indicate the copper wires were fully liberated from the polyester enamel in 24 h.

In summary, the results presented in Fig. 9 showed that, at room temperature, decreasing the amount of K₂CO₃ catalyst improves the DMT and DMI yield with the expense of a slower rate of reaction. Moreover, decreasing the DCM to methanol ratio did not increase the monomer yield but instead resulted in a much slower rate of reaction.

Thus, Run B in this study, with S/L, DCM/Methanol, and K₂CO₃/Cu_{wire} ratio of 500 g/L, 1.00 mol/mol and 0.10 wt%, respectively, as the optimum parameters for room temperature methanolysis.

The published research by Pham and Cho (2021) reveals that under the presence of moisture and K₂CO₃ catalyst, the DMT can partially hydrolysed to produce mono-methyl isophthalate or react with ethylene glycol to produce bis(2-hydro-xyethyl) terephthalate. Despite HPLC grade DCM and methanol being used in this study, they may still inherently contain small amounts of moisture of 0.05 wt% and 0.02 wt% respectively. It was suspected that the remaining 14.0% DMT and 7.8%DMI yield from Run B was due to the formation of by-products. To confirm this, the product from Run B was subjected to NMR analysis and the result is presented in supplementary information Fig. 7. The NMR analysis revealed the absence of bis(2-hydro-xyethyl) terephthalate by-products, and the products formed were indeed DMT, DMI and EG. Thus, the remaining 14.0% DMT and 7.8%DMI yield from Run B is due to loss during the recovery process.

Having shown that the methanolysis can be done at room temperature, the effect of increasing temperature was studied by varying the temperature from 25 °C to 35 °C while keeping other parameters the same as Run B and the results are presented in Fig. 10. From Fig. 8 it can be deduced that by increasing temperature from 25 °C to 35 °C, the rate of formation of DMT and DMI increases. At 35 °C, 87.8% DMT and 89.9% DMI yield can be achieved in 240 min. It is important to point out that the yield achieved in 240 min at 35 °C is almost equivalent to that of 25 °C in 24 h. Also, extending the reaction time at 35 °C to 24 h only led to a marginal increase in DMT and DMI yield of 87.9% and 91.1%, respectively which is within the standard deviation. Thus, at 35 °C, full decomposition can be achieved within 240 min. Furthermore, based on the recorded energy consumption from the hot plate used, the energy consumed for the complete decomposition of polyester enamel from 50 g enamelled copper wire at reaction temperatures of 25 °C and 35 °C were 0.22 kWh and 0.04 kWh, respectively. Hence, despite the higher temperature, the exponential increase in the rate of reaction from 25 °C to 35 °C with respect to temperature caused an overall energy savings of at least 5 times due to shorter reaction time from 24 h to 4 h. Furthermore, there is no gas by-product being produced by the proposed chemical method. In contrast, CO, CO₂, and harmful hydrocarbons are produced as by-products from the existing thermal treatment.

3.3. Comparison of the proposed process to pyrolysis

This subsection presents a simple energy consumption model to demonstrate the total electricity consumed for the proposed process compared to pyrolysis. The model assumes that the energy consumed varies linearly with the amount of waste enamelled copper wire being treated, such that the energy required to treat 1000 g of sample is equal to 20 times the energy required to treat 50 g of sample. In the calculation of the proposed process, it was assumed that the first

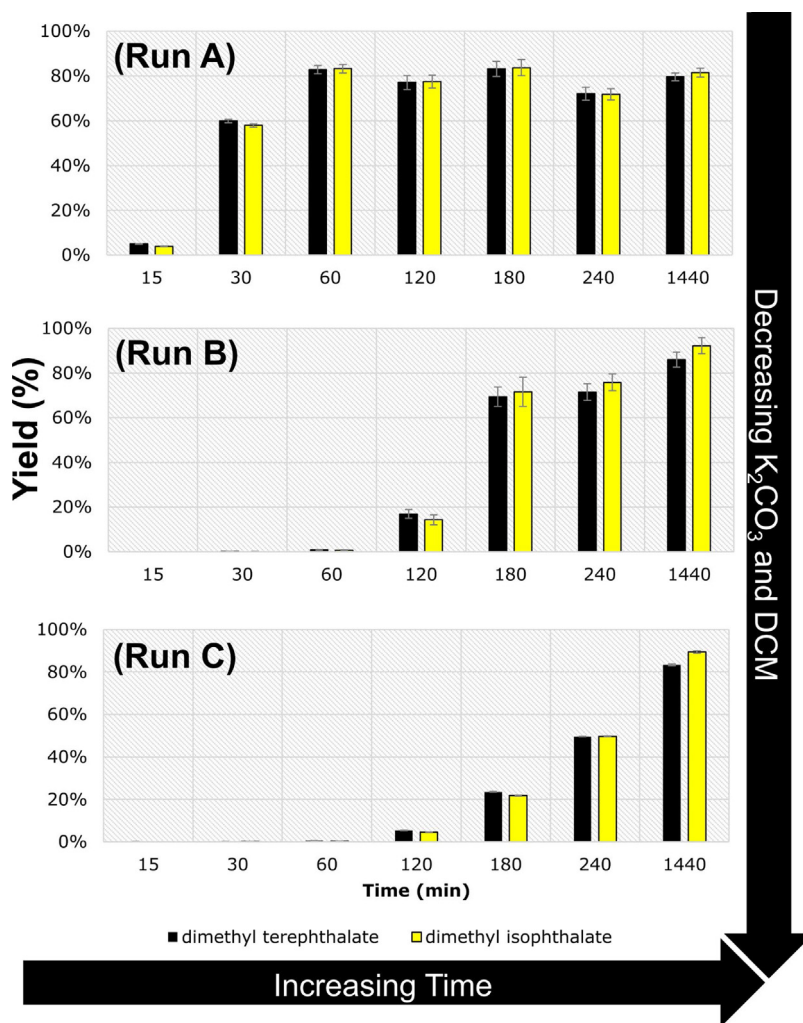


Fig. 9. Time versus dimethyl terephthalate and isophthalate yield for three different parameters of S/L, DCM/Methanol, and K_2CO_3/Cu_{wire} of;(Run A) 500 g/L, 1 mol/mol, and 7.5 wt%; (Run B) 500 g/L, 1 mol/mol, 0.10 wt%; (Run C) 500 g/L 0.2 mol/mol, 0.10 wt%. (the error bars are standard deviation).

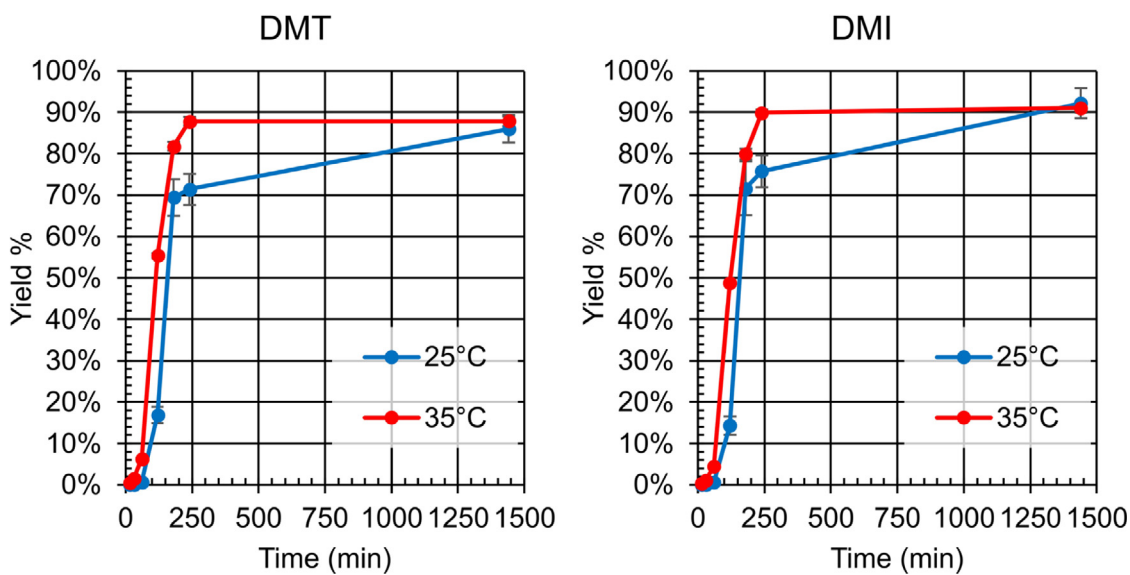


Fig. 10. Evolution of (left) DMT and (right) DMI from the methanolysis of polyester enamel at different temperatures. (Error bars are standard deviation).

Table 2
Total electricity cost of the proposed process and previously reported thermal treatment.

	Process	Unit Operation	Equipment	Rated Power (W)	Time (h)	Electricity Required (kWh)	Cost (£)	
Proposed Process	Methanol washing	Washing	Mixer	70	0.17	0.01	0.03	
		Methanol distillation	Rotavap	1400	0.50	0.70	0.18	
			Condenser	200	0.50	0.10	0.03	
			Vacuum pump	150	0.50	0.08	0.02	
	Methanolysis	Reactor	Hotplate stirrer (35°C)	/	4.0	(0.8) ^a	0.21	
		DCM and methanol distillation	Rotavap		1400	2.0	2.80	0.70
			Condenser		200	2.0	0.40	0.10
			Vacuum pump		150	2.0	0.30	0.08
	Total						5.19	1.35
	(Liu et al., 2020)	Thermal treatment	Pyrolysis	Furnace (warmup) 20°C – 600°C	5000	0.38	1.9	0.48
Furnace (pyrolysis) 600°C				5000	0.50	(25) ^a	6.25	
Total						26.9	6.73	

^a Basis of 1 kg enamelled copper wire.

stage of methanol washing was done in a single pass with a limit of 10 wt% and a residence time of 30 min. In the second stage, the methanolysis parameters of temperature, time, S/L, DCM/Methanol, and K_2CO_3/Cu_{wire} ratio of 35 °C, 4 h, 500 g/L, 1.00 mol/mol and 0.10 wt%, respectively, were used. The mass balance for the proposed process can be found in the supporting information. For the pyrolysis process, it was assumed that the pyrolysis only requires a single warmup stage. Moreover, for both processes, the furnace (pyrolysis), distillation, and washing unit operations always operate at full power due to the fluid that actively takes out heat from the system. The simple energy model results are presented in Table 2.

From Table 2, the total electricity required per kilogram of waste enamelled copper wire for the proposed process and previously reported pyrolysis process is 5.19 kWh and 26.9 kWh, respectively. The proposed process uses 80.7% less energy compared to pyrolysis, resulting in an overall operating cost of £1.35/kg compared to pyrolysis which costs £6.73/kg. Moreover, based on the current emission factor of 0.21 kg CO₂eq/kWh in the UK's national grid (ITPENERGISED, 2023), the proposed process releases 4.56 kg less CO₂ when compared to pyrolysis.

The proposed process shifts part of the cost load from electricity consumption to the chemical consumed. For reasonable comparison, the cost of chemicals consumed in the proposed process is as important as the cost of electricity consumed. The recent industrial prices of methanol and K_2CO_3 were 0.34 £/kg (BusinessAnalystIQ, 2023b) and 1.33 £/kg (BusinessAnalystIQ, 2023a), respectively. The methanolysis of 1 kg enamelled copper wire consumes 7.87 g of methanol which is equal to 0.27 pence. Moreover, 0.98 g of the K_2CO_3 catalyst can be assumed to be unrecovered and is equal to 0.13 pence. The total cost of methanol and K_2CO_3 per kg of enamelled copper can be calculated as £0.0030. Whereas the entire process consumes 5.19 kWh of electricity, equivalent to £1.35. Thus, the cost associated with the methanol and K_2CO_3 consumption is marginal when compared to the total electricity consumption cost.

Polyester manufacturing is energy intensive and highly polluting process (Palacios-Mateo et al., 2021). Based on the current manufacturing level, the embodied carbon of polyester is 32 kg CO₂ eq/kg (Kilgore, 2023). Each kilogram of copper wire is enamelled by 23.6 g of polyester, which translates to a potential saving of 0.8 kg CO₂ eq/kg. The proposed polyester treatment method not only saves electricity in the treatment process but also saves 0.8 kg CO₂ eq embodied carbon required to manufacture new polyester enamel per kilogram of copper wire. Moreover, the proposed method recycles the polyester which reduces the energy consumption in the new polyester manufacturing process. Thus, the proposed method has the potential to prevent 5.36 kg of CO₂ release per kg of waste enamelled copper wire when compared to thermal treatment.

4. Conclusion

The waste enamelled copper wire sample used in this study was found to be a multi layered type made up of polyester inner layer and PVB outer layer. A multistep process comprised of methanol washing and methanolysis was proposed. The PVB outer layer can be removed by dissolution using methanol, while the polyester enamel can be recovered in the form of dimethyl terephthalate and dimethyl isophthalate via room temperature methanolysis. Due to the relatively low PVB concentration (1.5 wt%) and thickness (5 µm), with a 1 to 1 weight methanol to waste enamelled copper wire ratio, a complete dissolution can be achieved in 1 min.

The results presented in this study demonstrate that decreasing the amount of K_2CO_3 catalyst improved the DMT and DMI yield with the expense of a slower rate of reaction while decreasing the DCM to methanol ratio did not increase the monomer yield but instead resulted in a slower rate of reaction. The optimum parameters for room temperature methanolysis were found to be S/L, DCM/Methanol, and K_2CO_3/Cu_{wire} ratio of 500 g/L, 1 mol/mol, and 0.10 wt%, respectively, with the reaction completed in 24 h. Increasing the temperature to

35 °C shortens the reaction time to 4 h and results in DMT and DMI yield of 87.8% and 89.9%, respectively, as well as lower overall reactor energy consumption.

A simple energy consumption model was used to assess the energy expenditure of the proposed process with solvent looping compared to the proposed thermal treatment found in the literature. It was found that the proposed process saved 80.7% of electricity which translates to 5 times lower electricity cost than the previously reported thermal treatment. The lower electricity usage and the recovery of the polymer as monomer prevented the release of 5.36 kg CO₂ eq per kilogram of waste enamelled copper wire. Moreover, there is no toxic gas formed from the proposed process. Hence, from the economic and environmental perspective, the proposed method is better than the existing thermal treatment.

The proposed method only considers one specific type of enamelled copper wire that is commonly used in electric motor and other types of enamelled may need different approach. Hence, this method requires prior segregation of enamelled copper wire to obtain the desired outcome and product. Compared to thermal treatment, the proposed process has lower productivity (i.e., 30 min pyrolysis vs. 4 h methanolysis), that may potentially require more space for scaling up. Moreover, despite the solvent used can be recovered and re-used, the K₂CO₃ catalyst has not been recovered. Future research direction should focus on other types of enamelled copper wire, recovery of used catalyst and whole system optimisation.

CRedit authorship contribution statement

Samuel D. Widijatmoko: Conceptualization, Visualization, Methodology, Investigation, Writing – original draft. **Zhehao (John) Cui:** Validation. **Hassan Agalit:** Validation. **Yongliang Li:** Resources, Supervision, Project administration, Funding acquisition. **Gary A. Leeke:** Supervision, Funding acquisition, Project administration, Writing – review & editing.

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.

Data availability

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

Acknowledgements

The authors would like to acknowledge funding from the Engineering and Physical Sciences Research Council (EPSRC), United Kingdom through the Plasma Assisted Thermo-Chemical energy storage for Carnot batteries (PATCH) Project: (EP/W027887/1).

The authors would like to thank European Metal Recycling Ltd. for providing the waste sample. The authors would also like to thank Luke Buckman for his technical support at the School of Chemical Engineering, University of Birmingham.

Appendix A. Supplementary data

Additional Supporting Information may be found in the online version of this article publisher's website.

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.resenv.2023.100143>.

References

- Ali, M.F., Hussain, M., Tariq, A., Ahmed, H.I., Shahid, S., Saouab, A., Nawab, Y., Sun, B., 2022. Damage-tolerant woven glass fiber composites developed using polyvinyl butyral (PVB) unsaturated polyester (UP) blends. *Adv. Mater. Sci. Eng.* 2022, 1–10.
- AquaSwitch, 2023. Business electricity prices.
- Barnard, E., Rubio Arias, J.J., Thielemans, W., 2021. Chemolytic depolymerisation of PET: a review. *Green Chem.* 23, 3765–3789.
- Bhanu, V.A., 2008. Polyester wire enamels - reliable coatings in the wire industry from the beginning to nowadays. *Macromol. Mater. Eng.* 293, 340–349.
- BusinessanalystIQ, 2023a. Methanol price index. Accessed 8 June 2023 (<https://businessanalytiq.com/procurementanalytics/index/methanol-price-index/>).
- BusinessanalystIQ, 2023b. Potassium carbonate price index. Accessed 8 June 2023 (<https://businessanalytiq.com/procurementanalytics/index/potassium-carbonate-price-index/>).
- Byrne, F.P., Jin, S., Paggiola, G., Petchey, T.H.M., Clark, J.H., Farmer, T.J., Hunt, A.J., Robert McElroy, C., Sherwood, J., 2016. Tools and techniques for solvent selection: green solvent selection guides. *Sustain. Chem. Process.* 4.
- Donelli, I., Freddi, G., Nierstrasz, V.A., Taddei, P., 2010. Surface structure and properties of poly(ethylene terephthalate) hydrolyzed by alkali and cutinase. *Polym. Degrad. Stab.* 95, 1542–1550.
- Elwert, T., Goldmann, D., Römer, F., Buchert, M., Merz, C., Schueler, D., Sutter, J., 2015. Current developments and challenges in the recycling of key components of (hybrid) electric vehicles. *Recycling* 1, 25–60.
- Essaddam, H., 2020. Polyethylene Terephthalate Depolymerization. Loop Industries Inc, U.S.
- Genta, M., Iwaya, T., Sasaki, M., Goto, M., Hirose, T., 2005. Depolymerization mechanism of poly (ethylene terephthalate) in supercritical methanol. *Ind. Eng. Chem. Res.* 44, 3894–3900.
- Haque, M.M., Kim, H., Kong, M.-S., Hong, H.-S., Lee, C.S., 2014. Optimizations of polyester degradation process of waste copper wire for the application of Cu recycling. *Polym. Eng. Sci.* 54, 607–615.
- Ioakeimidis, C., Fotopoulou, K.N., Karapanagioti, H.K., Geraga, M., Zeri, C., Papathanasiou, E., Galgani, F., Papatheodorou, G., 2016. The degradation potential of PET bottles in the marine environment: An ATR-FTIR based approach. *Sci. Rep.* 6, 23501.
- Ishihara, K., Ishida, K., Miyamoto, M., Nakashima, M., Sato, K., Hasegawa, H., 2004. Method for separating and recovering dimethyl terephthalate and ethylene glycol from polyester waste. Google Patents.
- ITPENERGISED, 2023. New UK grid emissions factors 2023.
- Jiang, J., Shi, K., Zhang, X., Yu, K., Zhang, H., He, J., Ju, Y., Liu, J., 2022. From plastic waste to wealth using chemical recycling: A review. *J. Environ. Chem. Eng.* 10.
- Kilgore, G., 2023. Carbon footprint of polyester vs cotton vs wool vs leather vs nylon (calculator). Accessed 10 August 2023 (<https://8billiontrees.com/carbon-offsets-credits/carbon-footprint-of-polyester/#:-:text=The%20carbon%20footprint%20of%20polyester%20can%20be%20explained%20as%20follows,driven%20by%20gas%2Dpowered%20car.>
- Li, B., Wang, X., Xia, Z., Zhou, W., Wu, Y., Zhu, G., 2023. Co-pyrolysis of waste polyester enameled wires and polyvinyl chloride: Evolved products and pyrolysis mechanism analysis. *J. Anal. Appl. Pyrolysis* 169.
- Liu, W., Wang, N., Han, J., Xu, J., Li, Z., Qin, W., 2020. Thermal degradation behaviors and evolved products analysis of polyester paint and waste enameled wires during pyrolysis. *Waste Manage.* 107, 82–90.
- Luan, W., Sun, L., Zeng, Z., Xue, W., 2023. Optimization of a polyvinyl butyral synthesis process based on response surface methodology and artificial neural network. *RSC Adv.* 13, 7682–7693.
- Ma, S., Xing, P., Li, H., Wang, C., Hou, X., Cun, Z., Liu, M., Yan, R., 2023. Recovery of high-grade copper from waste polyester imide enameled wires by pyrolysis and ultrasonic treatment. *Resour. Conserv. Recy.* 196.
- Miyamae, T., Yamada, Y., Uyama, H., Nozoye, H., 2001. Effect of oxygen radical and oxygen ion irradiation on biaxially stretched poly (ethylene terephthalate) surface studied by sum-frequency generation. *Appl. Surf. Sci.* 180, 126–137.
- Mominul Haque, M., Kim, H., Kong, M.-S., Hong, H.-S., Kim, K.-S., Lee, C.S., 2012. Recycling copper from waste copper wire using an applied voltage. *Int. J. Precis. Eng. Manuf.* 13, 1251–1254.
- Murray, T.J., 2008. Poly(amide-imides): Wire enamels with excellent thermal and chemical properties. *Macromol. Mater. Eng.* 293, 350–360.
- Nurdiawati, A., Agrawal, T.K., 2022. Creating a circular EV battery value chain: End-of-life strategies and future perspective. *Resour. Conserv. Recy.* 185.
- Palacios-Mateo, C., van der Meer, Y., Seide, G., 2021. Analysis of the polyester clothing value chain to identify key intervention points for sustainability. *Environ. Sci. Eur.* 33, 2.
- Paszun, D., Szychaj, T., 1997. Chemical recycling of poly (ethylene terephthalate). *Ind. Eng. Chem. Res.* 36, 1373–1383.
- Paul, C., Cotts, P., 1987. Static and dynamic light scattering from poly (vinyl butyral) solutions: effects of aggregation and solvent quality. *Macromolecules* 20, 1986–1991.
- Peer, P., Stenicka, M., Pavlinek, V., Filip, P., 2014. The storage stability of polyvinylbutyral solutions from an electrospinnability standpoint. *Polym. Degrad. Stab.* 105, 134–139.

- Petitgas, B., Seytre, G., Gain, O., Boiteux, G., Royaud, I., Serghei, A., Gimenez, A., Anton, A., 2011. High temperature aging of enameled copper wire—Relationships between chemical structure and electrical behavior. In: 2011 Annual Report Conference on Electrical Insulation and Dielectric Phenomena. IEEE, pp. 84–88.
- Pham, D.D., Cho, J., 2021. Low-energy catalytic methanolysis of poly(ethyleneterephthalate). *Green Chem.* 23, 511–525.
- Posavec, D., Müller, R., Bogner, U., Bernhardt, G., Knör, G., 2014. Polyvinyl butyral DMN-conjugates for the controlled release of singlet oxygen in medical and antimicrobial applications. *Biomater. Biomech. Bioeng.* 1, 73.
- Pudack, C., Stepanski, M., Fässler, P., 2020. PET recycling – contributions of crystallization to sustainability. *Chem. Ing. Tech.* 92, 452–458.
- Qin, X.-X., Cheng, Z.-L., 2016. Application of ionic liquids as a catalyst in the synthesis of polyvinyl butyral (PVB) polymer. *Chin. Chem. Lett.* 27, 145–148.
- Samide, A., Merisanu, C., Tutunaru, B., Iacobescu, G.E., 2020. Poly (vinyl butyral-co-vinyl alcohol-co-vinyl acetate) coating performance on copper corrosion in saline environment. *Molecules* 25.
- Scheirs, J., 1998. *Polymer Recycling: Science, Technology and Applications*. John! Wiley & Sons Ltd, Journals, Baffins Lane, Chichester, Sussex PO 19 1 UD, UK, p. 591, 1998.
- Shirazimoghaddam, S., Amin, I., Faria Albanese, J.A., Shiju, N.R., 2023. Chemical recycling of used PET by glycolysis using niobia-based catalysts. *ACS Eng. Au* 3, 37–44.
- Siddiqui, M.N., Redhwi, H.H., Achilias, D.S., 2012. Recycling of poly (ethylene terephthalate) waste through methanolic pyrolysis in a microwave reactor. *J. Anal. Appl. Pyrolysis* 98, 214–220.
- Tang, H., Su, Y., Hu, T., Liu, S., Mu, S., Xiao, L., 2014. Synergetic effect of LaB6 and ITO nanoparticles on optical properties and thermal stability of poly(vinylbutyral) nanocomposite films. *Appl. Phys. A* 117, 2127–2132.
- Ueda, K., 1989. Progress of magnet wire technology in Japan. *IEEE Electr. Insul. Mag.* 5, 18–26.
- Yang, B., Liu, R., Huang, J., Sun, H., 2013. Reverse dissolution as a route in the synthesis of poly(vinyl butyral) with high butyral contents. *Ind. Eng. Chem. Res.* 52, 7425–7431.
- Yang, Y., Lu, Y., Xiang, H., Xu, Y., Li, Y., 2002. Study on methanolytic depolymerization of PET with supercritical methanol for chemical recycling. *Polym. Degrad. Stab.* 75, 185–191.