Journal of Physics:
Conference Series

Mechanical and thermal characterization of a vitrimerized thermoplastic

To cite this article: Omar Amin *et al* 2025 *J. Phys.: Conf. Ser.* **3058** 012007

Mechanical and thermal characterization of a vitrimerized thermoplastic

Omar Amin [1], Wael S I Abou-Elmagd [2], Mohamed Hazem Abdellatif [1] and Amna Ramzy [3]

[1]Design and Production Department, Faculty of Engineering, Ain Shams University, Cairo, Egypt
[2] Chemistry Department, Faculty of Science, Ain Shams University, Cairo, Egypt
[3] Department of Materials Engineering, Faculty of Engineering and Materials Science, German University in Cairo, Main Entrance, El-Tagamoa Elkhames, New Cairo, Egypt
Corresponding author: omar.amin.mh@eng.asu.edu.eg

Abstract In this work, a vitrimer of polyethylene terephthalate was prepared through mixing with an epoxy and a polyol and curing. Different curing temperatures were utilized for preparation and the resulted characteristics were investigated. Curing temperature was found to highly affect the produced vitrimer. At high curing temperature, crosslinking was observed to be achieved successfully through different characterization methods. Differential scanning calorimetry DSC showed a single peak instead of dual peak known for PET. This proves disability of forming two populations of crystallites due to crosslinking. In addition, swelling test showed insoluble portion of the vitrimer, which indicated that crosslinking was obtained. Swelling in two different solvents were studied. On the other hand, at lower curing temperature, DSC showed dual peaks and tensile test showed lower tensile strength compared to precursor PET. Dynamic thermo-mechanical analysis did not show plateau after melting temperature, indicating non-networked structure.

Keywords: Vitrimers, Polyethylene terephthalate, Crosslinking, Characterization

1. Introduction

During processing, operating and recycling, plastics – for example Polyethylene terephthalate - lose their properties due to degradation. Chain extenders and stabilizers can be used to reclaim these properties; however, properties reduction will continuously occur during this cycle of re/processing and application. Chain extenders can also lead to branched and irreversible crosslinking which is non-processable. Solidstate polymerization is another solution; however, it has low efficiency. On the other hand, non-recycled polymers induce more microplastics into the environment and loss of resources. Around 10% of commercial polymers are polyesters [1]. Therefore, vitrimerized polyesters (e.g. Polyethylene terephthalate PET**)** offers a viable promising alternative, which sustains recyclability, simultaneously with enhanced properties accompanying the crosslinking. Different research groups addressed this point, and prepared PET vitrimers, through which chemical flexibility of upcycling approach was valorized. Most of their studies depended on using polyols'- or epoxies'- hydroxyl groups and a catalyst for branching. This was carried out by scissoring main chains of PET into oligomers and repolymerization into a branched crosslinked network [2].

Fabrizio et al [2] developed a vitrimer from melt blending of tray-PET and diepoxy of "Bis-phenol A", while incorporating Zinc acetylacetonate for transesterification catalysis. They used compression molding to develop test specimens. The procedure for the vitrimer preparation consisted of melt blending and compression molding. Future-recommended work was to study upcycling of textile PET. Joseph Ng et al [3] prepared a PET vitrimer by introducing glycerol to cleave and crosslink the chains, in addition to introducing Zinc catalyst for transesterification. While branching occurred after the addition of glycerol, crosslinking (gelation) on the other hand only started after the addition of and melt-blending with the Zinc catalyst. This was evidenced by crossover between dynamic moduli during rheology time-sweep test. The reason is probably because the catalyst promoted the transesterification after the reduction of abundant hydroxyl groups of glycerol which reacted with PET.

Qiu et al [4] used a procedure of melt-blending with a polyol and a diepoxy; to obtain oligomers and crosslinking, respectively. However, no catalyst was used. And afterwards curing and compression molding were employed to obtain the vitrimer samples. They utilized a tertiary amine polyol which provided internal catalysis and abundant hydroxyl groups, as alternatives for external catalyst for their PET-vitrimer. These alternatives are better from both the environmental and performance perspectives; as the external catalyst may leach out. However, the process required curing for several hours and specimens were prepared via compression molding, which is long, and non-industrial process. On the other hand, Li et al [5] provided a more industrial and greener approach by reducing synthesis time, and steps. PET vitrimer was developed by mixing with a dianhydride and a tertiary amine polyol. They used effects of Neighboring-group-participation in addition to catalysis reactivity of the anhydride with the hydroxyl, besides the tertiary amine and abundant hydroxyl groups for faster exchange reaction. This induces faster and continuous re/processing. The utilized procedure was melt-blending and compression molding. There are other studies in the literature which worked on enhancing properties of PET by vitrimerization and structure modification [6][7][8]

Previous work on PET vitrimer did not provide deep discussions about curing parameters and its relation to final properties or about the swelling test [2][3][4][5]. In this work, an experimental study of PET vitrimerization by adding a polyol and a diepoxy is presented. More insights are provided about the curing procedure and crosslinking behavior. Tensile properties are presented for dog-bone test specimens of vitrimerized PET and compared to control samples. Different approaches for swelling tests were discussed. More aspects are elaborated from results and thermographs of differential scanning calorimetry. Dynamic mechanical thermal analysis is performed and the results are discussed and reflected to other characterization outputs.

2. Experimental work

In this section, parameters of the tests are introduced, for tensile test, swelling test, DSC and DMA.

2.1. Materials
Spheroid pellets of recycled PET were provided by Bariq, Egypt. Polyethylene PE and Polypropylene PP were purchased from Sabic, Saudi Arabia. Diglycidyl ether of bisphenol A (DGEBA or BADGE) diepoxy, Irganox 1010 and 2(Bis(2-Hydroxyethyl) imino)-2-(Hydroxylmethyl)-1,3 Propanediol (Bis-Tris polyol) were obtained from Sigma-Aldrich. Trichloroacetic acid TCA was purchased from Algomhoria chemicals, Egypt. Trifluoroacetic acid TFA was acquired from Cornell Lab, Egypt. All chemicals were used as received.

2.2. Samples preparation
Three vitrimer samples (S1, S2, and S3) were prepared by mixing PET, epoxy, Bis-Tris and irganox 1010 in an extruder. Three control samples (CS1, CS2 and CS3) from only PET with irganox were prepared as well. Samples are cured at high temperature after that. The nomenclature is at the end of the paper (before the references). PE and PP were utilized to clean the extruder before and after our work.

2.3. Characterization methods
2.3.1. Tensile testing. Tensile testing was applied for injection molded specimens of CS2, CS3 and S3. Dog-bone test specimens were prepared according to ISO 527-1 and ISO 257-2 type A1. Specimen total length was approximately 150 mm and gauge length was taken as length between grips (115 mm). Crosshead speed was set at 5 m/min for all tensile tests. At first, tests for CS2 were implemented to study the injection molding parameters. The optimum results were sought for during the change of the parameters' values. Injection molding parameters included injection pressure, holding pressure, holding time, cooling time, plasticize quantity and temperatures of the barrel zones.

The resulted specimens were evaluated manually based on brittleness, strength, color (degradation), sink marks, wrapping, and cavities. In addition, clogging and leaking were avoided. In addition, for all injection molding specimens, first 3 or 4 cycles were avoided to ensure quality and consistency of the specimens. After that, tensile tests were implemented for dog-bone samples "S3B2cured200-15" and CS3.

2.3.2. Swelling test. Different approaches were used, starting with putting the samples S1 and PET pellets in TCA solvent for 4 days at room temperature. The other approach is by refluxing in TCA for 24hrs, for same samples. TFA is used after that for S1, S2 and CS1 samples at room temperature. The used samples were of approximately 0.35 to 0.55 mg and arbitrary geometry or as pellets.

2.3.3. Differential scanning calorimetry DSC. DSC test was performed with a TA Q2000 from TA instruments as follows. Heat/cool/heat cycles were implemented from 25°C to 300°C to 25°C again using a rate of 10°C/min for the ramping up and down. Three isothermal minutes at 300 C was done to erase thermal history for the samples. Purge gas was Nitrogen with 50 mL/min flow rate.

2.3.4. Dynamic mechanical thermal analysis DMA. DMA was performed with a DMA Q800 from TA instruments, in a tensile mode and following standard ASTM D5026. Length was ~ 10 to 15 mm, width: 6 to 10 mm, and thickness: < 2 mm. The used parameters were: 25 C to 300°C range, 5°C/min rate, 1 Hz frequency, 4 μm amplitude, in addition to air environment.

2.3.5. Morphology of filaments. Filaments were produced from PET and PET vitrimer pellets. Using an optical microscope, the surface texture of the filaments was observed. And using a vernier caliper the uniformity of the diameter was checked at 15 different positions along the filaments.

3. Results and discussion

In this section, curing results for different samples at different conditions are introduced and discussed. After that, tensile testing results are illustrated. Swelling test outcomes are presented afterwards. Finally, DSC and DMA graphs are shown.

3.1. Curing

3.1.1. Curing of Sample S1. Qiu et al [4] reported a successful postcuring under 205ºC for 10 hrs and nearly successful postcuring under 240ºC for 7 hrs. Different curing temperature and duration values recommended were reported in [4] and [9]. Interpolated durations for the selected temperature were used in our study as well be shown. In [9], PET was melt blended with epoxies from the same family as BADGE epoxy used in our study. They reported improvements in the properties of injection-molded sample if heat-treated under the recommended range of temperatures and durations.

Therefore, our vitrimer sample S1 was heated in an oven at a range of 230 to 250ºC for 7 hrs. The sample S1 was slightly melted and oxidized. The sample was noticed to still being brittle. At the same time, PET pellets were put in the same oven for the same temperature and time. Small portion of the pellets' surface color changed into light orange. This is due to oxidation. However, the pellets were not melted. This may indicate that the vitrimer had lower melting point than the pellets, which matches with the results by Qiu et al [4]. This may be because those fresh pellets were not exposed to polymer processing which causes degradation in addition to alcoholysis due to reaction with the polyol.

It can be concluded that these PET pellets and the vitrimer sample have melting points at around 250ºC to 260ºC and 245ºC to 250ºC, respectively. This will be further studied by DSC. In addition, it can be noticed that the oxidation rate is very high after melting. Further curing processes for similar vitrimer samples was recommended to be under 220ºC to 230ºC to avoid melting and consequently higher oxidation. In addition, it was recommended to use a more accurate oven.

To decrease the temperature, the postcuring time should be increased [10]. Using interpolation, the chosen temperature 220ºC is supposed to require approximately 8.75 hrs. Subsequently, Vitrimer sample S1 was heated under 220 to 240ºC for 8.5 hrs, figure 1. The oxidation level is noticed to be lower than the previous samples. In addition, the sample showed lower brittleness under a simple compressive-failure-test using the workshop vice. The fractured particles showed a white inner surface which mean that the oxidation was mainly on the sample surface.

Figure 1. Vitrimer S1 B1 after heating under 220 to 240ºC for 8.5 hrs

3.1.2. Curing for sample S2. Sample S2 were post-cured using the successful approach used for S1. The samples were heated for 8.5 hrs under 210 to 240°C.

3.1.3. Curing of sample S3. After pelletizing, S3 pellets were dried in a dehumidifier under 130°C for 3 hrs as implemented in a patent before [9]. S3 pellets were post-cured for 8 hrs under 120 to 200°C in the dehumidifier (Maximum temperature of the dehumidifier). The eight-hours duration was the maximum continuous duration for using the dehumidifier. Subsequently, small portion of the sample "S3B2cured200-8" pellets were extruded in a small extruder to examine its extrudability and flowability after post-curing. Afterwards, the sample "S3B2cured200-8" was cured for more 7 hours and those pellets were used to prepare injection-molded tensile samples. Other S3 pellets were post-cured under 220°C for 8.5 hrs in a Muffle furnace.

3.1.4. Curing of samples CS1 and CS3. Post-curing of sample CS1 was implemented for 8.5 hrs under 220°C. CS3 sample was cured in the dehumidifier for 15 hrs at 120 to 200°C. This is to enable to compare it with "S3B2cured200-15" sample.

3.2. Tensile test results

CS2I specimens were the first specimens to be prepared using injection molding in the project. Therefore, injection parameters were being changed and varied for optimization and consequently the specimens had a lot of defects. Results of tensile testing CS2 show an average tensile strength of below 20 MPa. The optimized injection parameters were used after that, table 1.

Table 1. Values for injection parameters of tensile specimens

Injection parameter (unit)	Value
Injection pressure (bar)	50
Holding pressure (bar)	45
Clamping pressure (bar)	85
Holding time (s)	5
Cooling time (s)	10 or 25
Temperatures of four barrel-zones (°C)	245, 250, 255, 260 (nozzle zone) Or 250, 260, 270, 280 (nozzle zone)
Plasticize quantity (mm)	55

For S3 and CS3 samples, the stress-strain curve is presented in figure 2. Number of specimens is seven for S3 and three for CS3. Lower strength of the vitrimer could be due to alcoholysis, lower crosslinking, degradation during processing or non-sufficient drying. Lower tensile properties could be due to lower crystallinity after crosslinking as well (competing factors effect [11]). Fabrizio et al [2] suggested that the stiffness is only slightly influenced by crosslinking and highly affected by crystallinity. Tensile modulus was reported to be lower for the vitrimerized PET as compared to the precursor PET, and this was ascribed to the lower crystallinity. However, the tensile modulus for our work was the same for both specimens or slightly higher for the vitrimer. It should be noted here that during producing the vitrimer tensile-test samples on the injection molding machine, the nozzle was clogged most of the time after injecting and ejection of the samples. This was mostly due to high adhesivity of the vitrimer, especially that it contains epoxy, and solidification of the vitrimer at the nozzle after that.

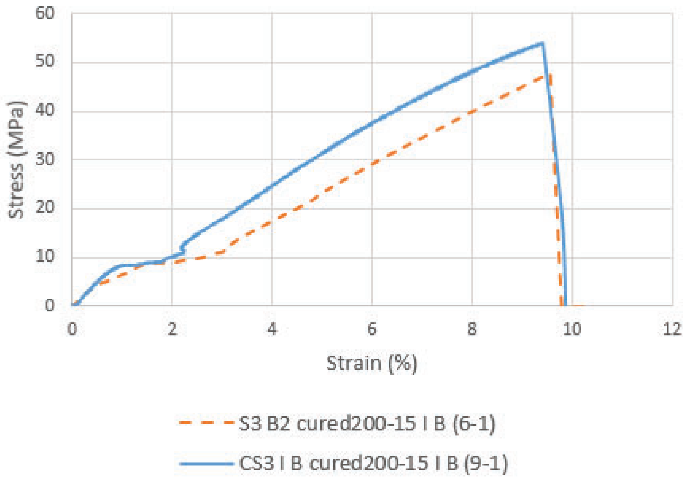


Figure 2. Stress strain curves for S3B2cured200-15 I vs CS3Bcured200-15 I

3.3. Swelling test

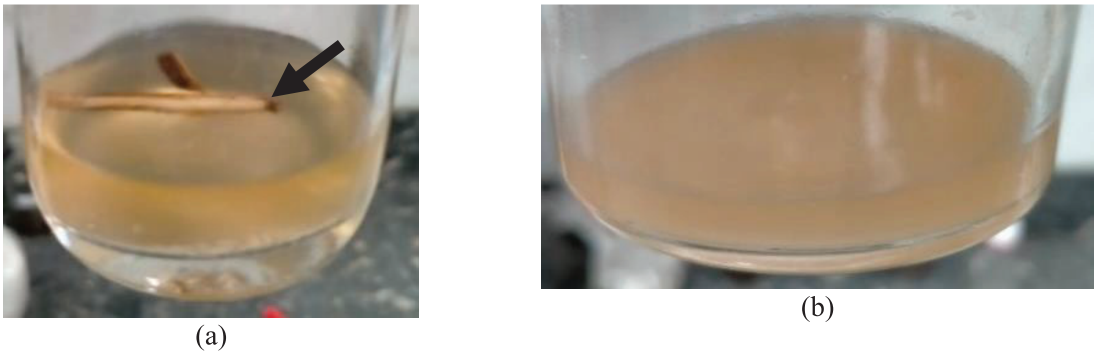
Due to the low availability in local market, of the solvents demonstrated by other PET vitrimer studies [3][4][5] like (1,1,1,3,3,3-hexafluoro-2-propanol (HFiP) and (TFA), it was suggested to look up for other solvents for PET in literature. It was found that Trichloroacetic acid (TCA) is a solvent for PET as well and it was found to have lower cost than the TFA, and higher availability [12]. Therefore, it was suggested to use it as a first trial to dissolve PET and for swelling test.

To prepare a 100 w/v% solution of TCA, a method was adopted from Sigma-Aldrich. To prepare the solution, 88.1 gm of solid TCA was added to 25 ml distilled water. After the addition and stirring, the final volume of the solution was 80 ml. Two 100 ml beakers were used to contain equal quantities (20 ml) of the solution. A 0.5 gm of both PET and Vitrimer were added to the beakers separately with some stirring. After 4 days at room temperature, neither PET nor Vitrimer were dissolved. Therefore, it was proposed to use another approach through heating for 24 hrs or to utilize the TFA instead of TCA.

The 100 w/v% solution of TCA was prepared by adding 88.1 gm of TCA to tap water and stirring. The volume of the solution at the end is approximately 100 ml. About 0.5 gm of PET pellets were put in the acid in 100 mL round bottom flask and refluxed on a hot plate at around 150°C with a condensing mechanism. After 5 hrs of refluxing (implemented in different days), the pellets turned into powder, and after 14 hrs and 48 hrs, it remains as powder as well. This could be due to the acidic hydrolysis of PET into Terephthalic acid (powdery precipitates) and ethylene glycol (yellow liquid) [13].

Triflouroacetic acid TFA was used and the chosen approach was through putting the sample in the acid at room temperature. This is a safer approach due to the higher danger of TFA compared to TCA and compared to other acids. Firstly, a control experiment/test was implemented using PET pellets to ensure its solubility in TFA. A 20 mL of TFA (synthesis grade, with minimum assay of 99%) was put in a 100 mL beaker and a 0.5 gm of PET pellets were put in it and stirred a little. The beaker was covered with a petridish to reduce the danger and solvent loss from TFA's fumes. It was noticed that TFA has continuous fumes. Therefore, it is recommended to be used under a fuming hood, especially when heated. After 48 hours, the pellets were checked and were noticed to be dissolved, and the color of the solvent was a bright orange. The level of the solvent decreased due to the continuous fumes. Therefore, it is recommended to implement this test in a more tightly closed container.

3.3.1. Swelling of sample CS1. Swelling test was carried out for a 0.5 gm of the post-cured CS1 in approximately 10 mL of TFA. The solvent was contained in glass bottle with screwed plastic cap. The solvent was filtered after 4 days to check the characteristics of the residues. The filtered residues consisted only of small black pieces which are possibly the oxidized outer layer of the post-cured control sample 1 (CS1). The filtered acid was kept to be used for another swelling test, figure 3.


(b)

(a)

Figure 3. Swelling test for CS1 (a) at the test start (b) Filtered acid

3.3.2. Swelling of Sample S1. A small quantity of TFA of around 25 mL was utilized to implement swelling test for S1 sample (before curing). A sample of 0.35 gm weight was put in the TFA solvent in a glass container with a glass cap to keep reduce solvent losses due to evaporation. After 24 hrs, there was still a solid part of the sample which vanished after little stirring. However, at the end of stirring, there was still some fibrous residues. This could be due to some level of cross linking of the vitrimer inside the extruder.

After that, a post cured vitrimer sample S1B1cured220-8 (cured at 220^{o}C for 8 hrs) was put in the same solvent quantity. After 72 hrs, the sample converted into fibrous/gelly pieces, figure 4.


(a) (b)

Figure 4 Post cured vitrimer sample S1B1cured220-8 in TFA (a) at the test start (b) swelling of sample after 72 hrs.

3.3.3. Swelling of sample S2. For S2 sample, swelling test was implemented as follows. An approximately 25 mL of TFA in a 250-mL glass container with glass cap was utilized. It was implemented for the S2 before curing (0.55 gm) and after curing (0.45 gm). Before post-curing, the sample dissolved completely in TFA. After post-curing, there is noticed residues after 48 hr and 4 days. The residues have brown to black color which indicates that the residues could be only the outer surface of the post-cured sample. The residues got pale colors and more transparent appearance when checked after 4, 5, 7, 10, 13 and 15 days. The swelling of the PET vitrimer and dissolution of its precursor PET was reported as well by Qiu et al [4]. However, after more than two months only contaminants appear in the solution. It was reported that vitrimers can completely or partially dissolve. This depends on several parameters among which are the network topology and experiment time [14].

3.4. DSC

The 2nd heating scans were checked in our study to investigate the changes in the material structure. Alcoholysis of the non-cured vitrimer is reflected in the lower melting points Tm and glass transition temperature Tg compared to control sample CS3. Tm for the non-cured vitrimer is 240.6°C compared to 248°C for the control sample. Tg for the non-cured vitrimer is 76.7°C compared to 79.8°C for the control sample. This is due to more flexible shorter chains. Crosslinking of vitrimer samples can be illustrated with the higher Tg and lower Tm and melting enthalpy (due to lower crystallinity) compared to the non-cured vitrimer samples, figure 5. Tm for the vitrimer sample is 238.7°C compared to 240.8°C for the non-cured vitrimer. Tg for the vitrimer is around 82°C compared to 78.2°C for the non-cured vitrimer. The crosslinking can be concluded as well from single melting peak of the cured vitrimer sample compared to the dual-melting-peaks of the non-cured samples, figure 5. This shows an agreement with other studies in the literature which demonstrates that the cross linking reduces crystallinity due to difficult chain packing [2]. Crosslinking in our experiments induced lower Tm and lower ΔHm, in agreement with the research work of Fabrizio et al [2]. In addition, DSC from study by Li et al [5], demonstrates single melting peak after vitrimerization of PET, as compared to dual melting peak of the PET precursor.

Another cured vitrimr sample S3 B2cured200-8, which is cured at range under 200°C in a dehumedifier, showed increased Tg compared to the non-cured S3B2. However, its Tg is lower than the sample cured at 220°C. As shown in the figures, Tg for the non-cured sample is 78.2°C, while for the sample cured at 200°C it is 79.2°C, and for the sample cured at 220°C is 81.9°C. This is despite the longer curing time (15 hrs) in the case of 200°C compared to 8 hrs for the sample cured at 220°C. It should be noted here that heat treatment of the PET at this range of temperature is cosidered as annealing which can affect the apparent properties even if it is not affecting the chemical structure. This annealing is an aging process which was reported for PET in literature as well [15][16].

3.5. DMA

As shown in figure 6, the Tm in the DMA for the vitrimer was found to be lower than the control sample CS3I. However, the storage modulus for the vitrimer is higher. On the other hand, the vitrimer's characteristic rubbery plateau after melting did not appear. This is similar to the result demonstrated by Fabrizio et al for PET-vitrimer with lowest crosslinking [2]. This shows that the vitrimer cured at lower temperature didn't reach a sufficient crosslinking level. This could be due to degradation as well.

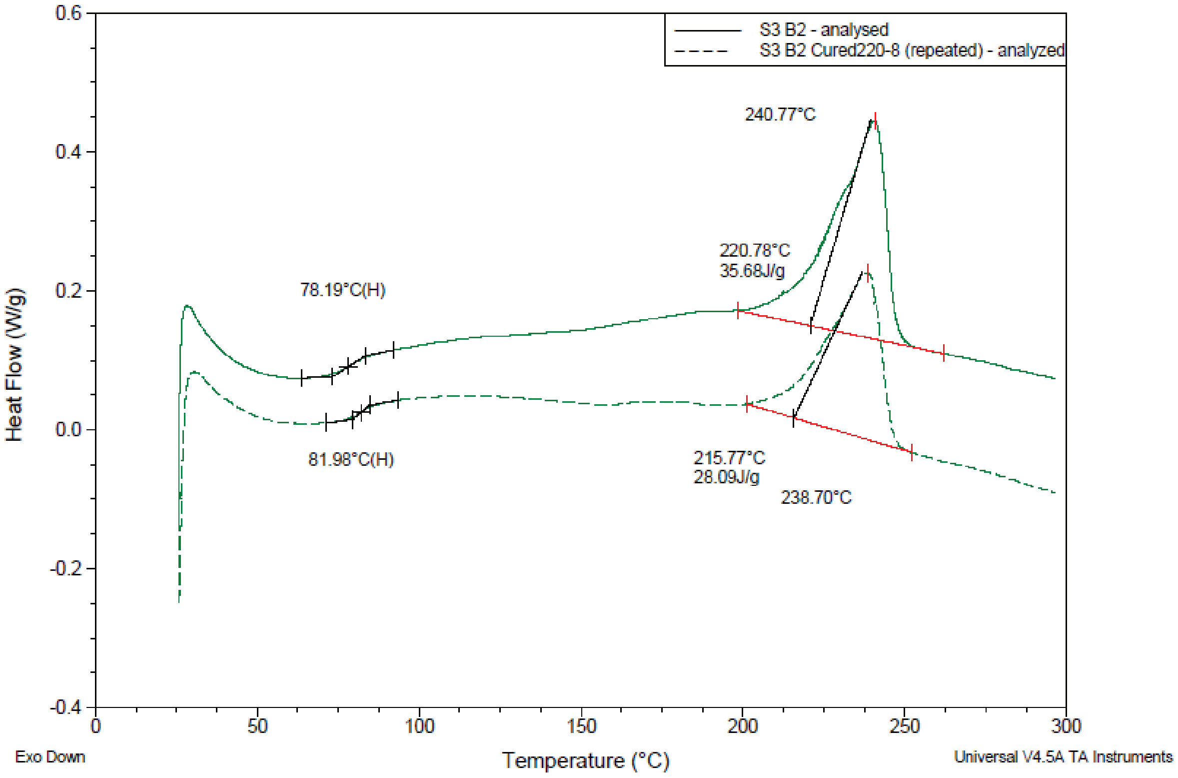


Figure 5. Crosslinking effect

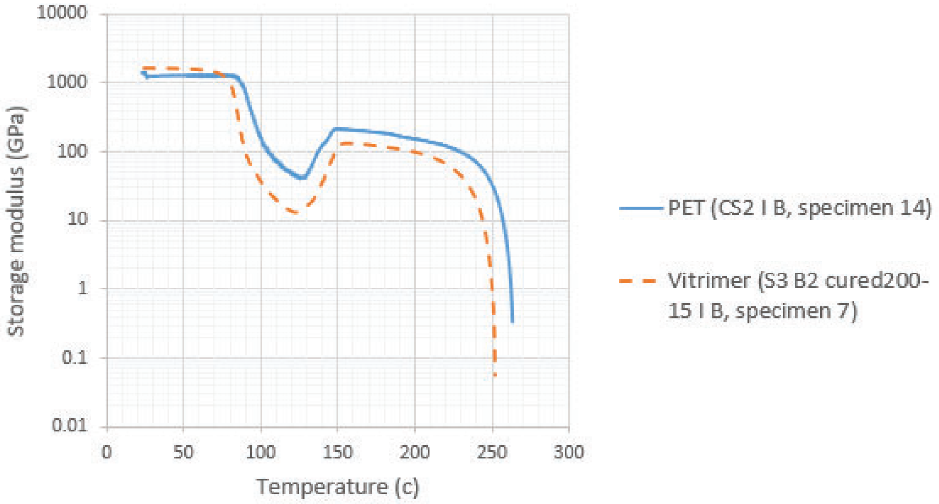


Figure 6. DMA storage modulus curves for S3B2cured200-15 vs CS2 I B (as control sample)

3.6. Morphology of filaments

Using an optical microscope, it was noticed that the vitrimer filaments had smoother surface when compared to PET filaments, figure 7. In addition, the diameter variations for the vitrimer filament were lower than the PET filaments, figure 8. It should be noted that this vitrimer filament is produced from the pellets cured under temperatures lower than 200ºC. These observations could result from lower viscosity of epoxy and PET after alcoholysis and subsequently higher self-leveling [17][18].

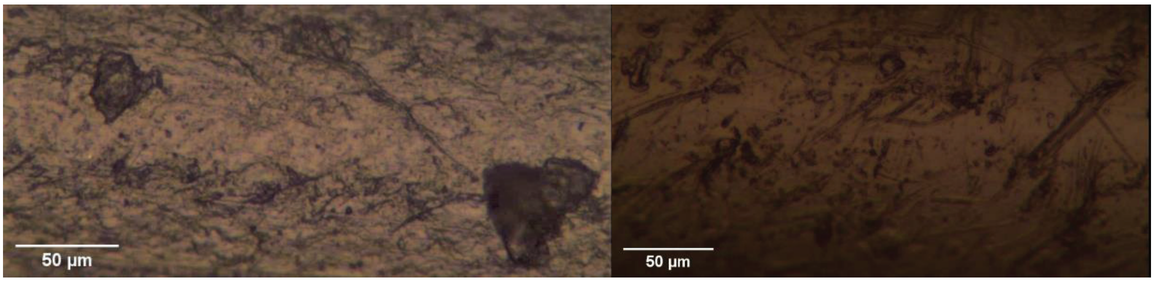


Figure 7. Morphology of filaments (left) PET, (right) vitrimer

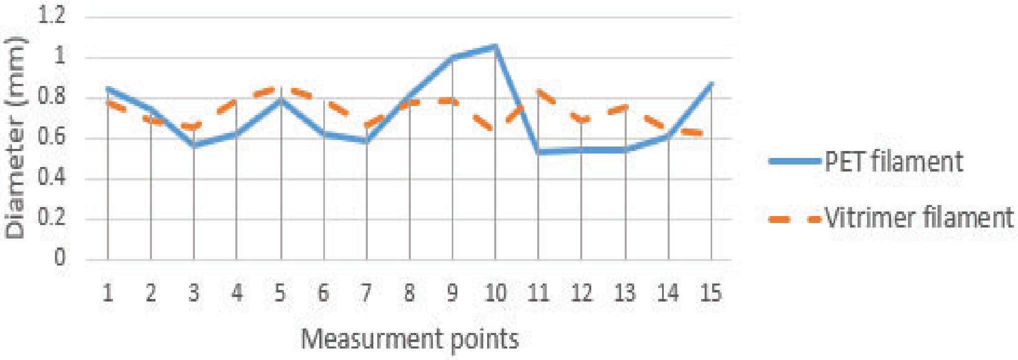


Figure 8. Filaments' diameters

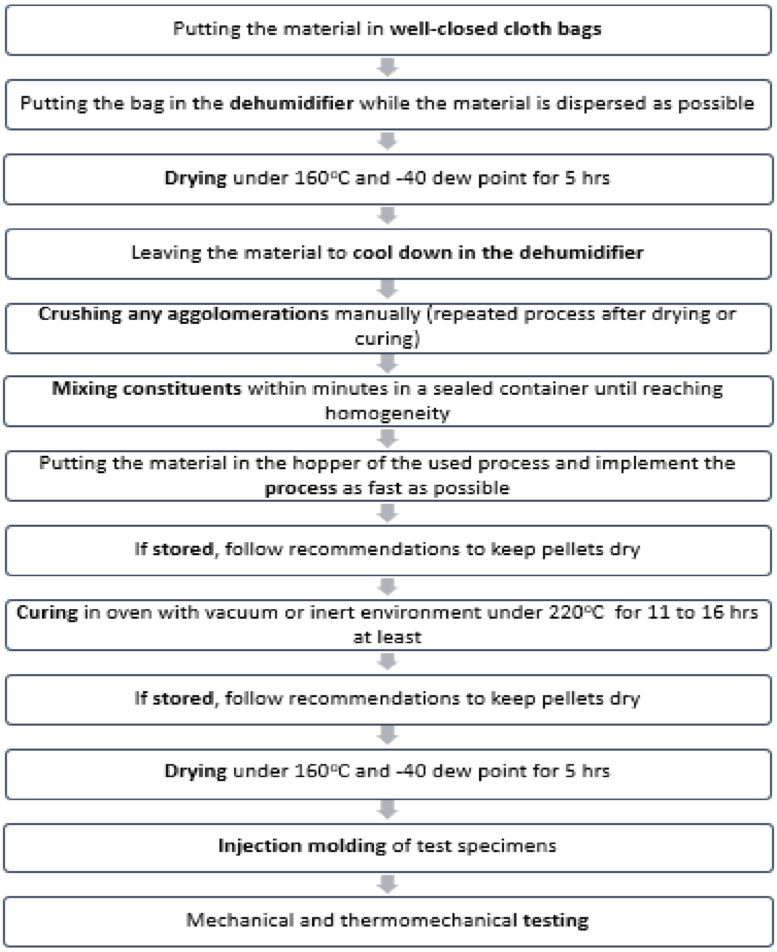
4. Conclusions

Crosslinking and vitrimerization of PET-blend was achieved through curing at temperatures in a range from 120ºC to 250ºC. At certain chosen high curing temperatures (around 220ºC), the crosslinking was successful. This was proved by DSC through lower Tm, higher Tg and single melting peak. This was evidenced as well using swelling test, where vitrimer samples converted into jelly substance and didn't fully dissolve. Dissolution in TCA was shown to be very slow and impractical. The most suitable and economic swelling test was presented by TFA at room temperature for a day to 4 days. At lower curing temperatures (in the range of 120ºC to 200ºC), the crosslinking was not successful. This was shown by the lower tensile strength and non-appearance of rubbery plateau in the DMA. In addition, this method for producing PET vitrimer provides smoother surface texture compared to PET. Moreover, it is recommended to use compression rather than injection molding for this vitrimer due to the nozzle clogging issue, especially that it is still in its initial research stages.

PET is usually used for (i) food (and other substance) packaging, (ii) fibers for clothes, textiles, carpets and automotive tires), (iii) films and (iv) components for electronic and automotive industry. PET vitrimer can be used in the applications requiring chemical resistivity. In addition, it should provide better recyclability performance; i.e. better properties for the recycled parts and better mechanical properties. This needs further investigations. Nevertheless, in case of using "bisphenol A" to transfer PET into vitrimer, its use for food and beverages packaging may be less safe.

5. Recommendations and Future work

The recommended procedure for sample preparation – based on our results - is illustrated in figure 9.



Putting the material in **well-closed cloth bags**
Putting the bag in the **dehumidifier** while the material is dispersed as possible
Drying under 160ºC and -40 dew point for 5 hrs
Leaving the material to **cool down in the dehumidifier**
Crushing any aggolomerations manually (repeated process after drying or curing)
Mixing constituents within minutes in a sealed container until reaching homogeneity
Putting the material in the hopper of the used process and implement the **process as fast as possible**
If **stored**, follow recommendations to keep pellets dry
Curing in oven with vacuum or inert environment under 220ºC for 11 to 16 hrs at least
If **stored**, follow recommendations to keep pellets dry
Drying under 160ºC and -40 dew point for 5 hrs
Injection molding of test specimens
Mechanical and thermomechanical **testing**

Figure 9. Recommended experimental work procedure

In addition, it is recommended for future studies to implement mechanical characterization for this PET vitrimer after recycling. This is addition to energy consumption, environmental impact studies and long-term stability tests. This will ensure its sustainability and increase its chances to be adopted by research and industrial communities. Moreover, it is important to consider competing factors' effect when the structure is changed and manipulated [11]. Furthermore, long curing durations should be carefully considered because aging may alter the polymer's properties [15][16].

Statements and Declarations

Acknowledgement This work has been supported by the "3D Vitri-PET" Project funded by the German Academic Exchange Service (DAAD) and the Federal Ministry for Education and Research. Funded by: BMBF-/DAAD-TNB Bina Research Cluster under project ID: BMBF 57441946.

Authors' Contributions Conceptualization, Funding acquisition, Resources, and Project administration were performed by Dr. Amna Ramzy. Investigation, Formal analysis, Data curation, Visualization, Writing – Original Draft were performed by Omar Amin. Validation was performed by Dr. Amna Ramzy and Omar Amin. Methodology was performed by Dr. Amna Ramzy and Dr. Wael S. I. Abou-Elmagd. Review was performed by Dr. Amna Ramzy and Dr. Mohamed Hazem Abdellatif. Supervision was performed by Dr. Amna Ramzy, Dr. Wael S. I. Abou-Elmagd, and Dr. Mohamed Hazem Abdellatif

Data Availability Statement The data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflict of Interest The authors declare no conflict of interest.

Nomenclature

Code	Nomenclature
CS1	Control sample 1 which was produced by simply extruding and pelletizing PET pellets. As all other samples, it was produced from a six-zones twin-screw extruder
CS2	The same as CS1, however, with different extrusion conditions.
CS3	The same as CS2, however, with adding Irganox 1010 during extrusion.
S1	A vitrimer sample. It is the only sample in the study to be produced from a four-zones single screw extruder
S2	A vitrimer sample. This sample had high porosity due to inefficient drying.
S3	The same as S2, however, with better drying and mostly no porosity.
S3B2cured220-8	S3 sample cured under around 220C for 8 hours. The letter B after the first term indicates a certain batch of the main sample. Examples: S3B2 is a sample B2 of the S3; where B2 was the batch with the highest quality amongst all S3 samples.
CS2I	The "I" here indicates injection molding process for preparing specimens.

References

[1] Kimura T and Hayashi M 2022 One-shot transformation of ordinary polyesters into vitrimers: decomposition-triggered cross-linking and assistance of dynamic covalent bonds. *J Mater Chem A* ;**10**:17406–14. https://doi.org/10.1039/d2ta04110c.

[2] Fabrizio L, Arrigo R, Scrivani MT, Monti M and Fina A 2022 Upcycling of PET from recycled food packaging trays via vitrimers chemistry. *Polymer (Guildf)*;**266**:125618. https://doi.org/10.1016/j.polymer.2022.125618.

[3] Ng KWJ, Lim JSK, Gupta N, Dong BX, Hu CP and Hu J et al 2023 A facile alternative strategy of upcycling mixed plastic waste into vitrimers. *Commun Chem* ;**6**:1–12. https://doi.org/10.1038/s42004-023-00949-8.

[4] Qiu J, Ma S, Wang S, Tang Z, Li Q, Tian A, et al 2021 Upcycling of Polyethylene Terephthalate to Continuously Reprocessable Vitrimers through Reactive Extrusion. *Macromolecules*;**54**:703–12. https://doi.org/10.1021/acs.macromol.0c02359.

[5] Li P, Lan B, Zhang X, Lei S, Yang Q, Gong P, et al 2022 Facile in situ construction of a covalent adaptable network polyester vitrimer with advanced performance in repairability, foamability and recyclability. *Green Chem*;**24**:5490–501. https://doi.org/10.1039/d2gc01320g.

[6] Wu S-S, Li Y-D, Hu Z and Zeng J-B 2023 Fast Upcycling of Poly(ethylene terephthalate) into Catalyst-free Vitrimers. *ACS Sustain Chem Eng*;**11**:1974–1984.

[7] Chen CW, Mao HI, Lee HK, Chou JY and Rwei SP 2023 Upcycling Recycled Epoxy-Based Vitrimer for Enhancing Toughness of Poly(ethylene terephthalate). *J Polym Environ*;**32**:621–31. https://doi.org/10.1007/s10924-023-02996-7.

[8] Raffa P, Coltelli MB and Castelvetro V 2014 Expanding the application field of post-consumer poly(ethylene terephthalate) through structural modification by reactive blending. *J Appl Polym Sci*;**131**:1–11. https://doi.org/10.1002/app.40881.

[9] Siggel E, Rein SW, Obernburg, Koepp H-M, Erlenbach, AG G, et al 1971 Polyethylene terephthalate injection molding compositions containing a polyepoxide. US3560605A (Patent).

[10] Epoxy Chemistry n.d. www.westsystem.com/instruction/epoxy-basics/epoxy-chemistry/ (accessed May 21, 2022).

[11] Amin O, Ramzy A, Abou-Elmagd WSI and Abdellatif MH 2025 Enhanced healability and weldability properties of vitrimers : Review article. *Polym Eng Sci*;**65**. https://doi.org/10.1002/pen.27086.

[12] Mahalingam S, Raimi-Abraham BT, Craig DQM and Edirisinghe M 2015 Solubility-spinnability map and model for the preparation of fibres of polyethylene (terephthalate) using gyration and pressure. *Chem Eng J*;**280**:344–53. https://doi.org/10.1016/j.cej.2015.05.114.

[13] Han M 2019 Depolymerization of PET Bottle via Methanolysis and Hydrolysis *Recycling of Polyethylene Terephthalate Bottles* (vol 1) ed Thomas S, Rane A et al (Amsterdam: Elsevier Inc.) chapter 5 pp 85-108 https://doi.org/10.1016/b978-0-12-811361-5.00005-5.

[14] Van Zee NJ and Nicolaÿ R 2020 Vitrimers: Permanently crosslinked polymers with dynamic network topology. *Prog Polym Sci*;**104**:1–37. https://doi.org/10.1016/j.progpolymsci.2020.101233.

[15] Panowicz R, Konarzewski M, Durejko T, Szala M and Łazi M 2021 Properties of Polyethylene Terephthalate (PET) after Thermo-Oxidative Aging. *Materials (Basel)*;**14**:3833. https://doi.org/https://doi.org/10.3390/ma14143833.

[16] Farhoodi M, Mousavi SM, Sotudeh-Gharebagh R, Emam-Djomeh Z, Oromiehie A and Mansour H 2012 A study on physical aging of semicrystalline polyethylene terephthalate below the glass transition point. *J Appl Res Technol*;**10**:698–702. https://doi.org/10.22201/icat.16656423.2012.10.5.360.

[17] Khoshnevis H and Zadhoush A 2012 The influence of epoxy resin on the morphological and rheological properties of PET/PA66 blend. *Rheol Acta*;**51**:467–80. https://doi.org/10.1007/s00397-011-0615-5.

[18] Saalbrink A, Mureau M and Peijs T 1997 Blends of jcpoly(ethylene terephthalate) and epoxy as matrix material for continuous fibre reinforced composites. *Plast Rubber Compos Process Appl*;**30**:213–21. https://doi.org/10.1179/146580101101541642.