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jmr&t Journal of Materials Research and Technology

journal homepage: www.elsevier.com/locate/jmrt

Review Article

A review on the potential of polylactic acid based thermoplastic elastomer as filament material for fused deposition modelling

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ARTICLE INFO

Article history:
Received 19 May 2022
Accepted 11 August 2022
Available online 18 August 2022

Keywords:
3D printing
Additive manufacturing
Fused deposition modelling
Thermoplastic elastomer
Natural rubber

ABSTRACT

Currently, a range of sectors are implementing three-dimensional (3D) printing, which is a part of additive manufacturing (AM) technology via the fused deposition modelling (FDM) approach. As of now, various filament materials are available in the market and have their limitations. Thermoplastic elastomer (TPE) blend as a filament material in 3D printing should be implemented to overcome the weakness of available filaments. TPE blend stands out due to its flexibility, thermoplastic-like processability, and renewability. Based on the findings, TPE blend filament can be made with polylactic acid (PLA) thermoplastic and elastomers such as natural rubber (NR) and epoxidized natural rubber (ENR). The TPE printed components will be flexible; tough with excellent thermal and mechanical properties. In this paper, the characteristics of TPE are being reviewed to show the potential of TPE material as filament.

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https://doi.org/10.1016/j.jmrt.2022.08.057

List of abbreviations

ABS	Acrylonitrile butadiene styrene
AM	Additive manufacturing
CAD	Computer-aided design
DCP	Dicumyl peroxide
ENR	Epoxidized natural rubber
ESO	Epoxidized soy oil
FDM	Fused deposition modelling
FFF	Fused filament fabrication
Tg	Glass transition temperature
GMA	Glycidyl methacrylate
LENR	Liquid epoxidized natural rubber
LDPE	Low-density polypropylene
MA	Maleic anhydride
MFI	Melt flow index
Tm	Melting temperature
NR	Natural rubber
PAA	Poly (acrylic acid)
PTT	Poly (trimethylene terephthalate)
BR	Polybutadiene rubber
PBAT	Polybutylene adipate terephthalate
PCL	Polycaprolactone
PEEK	Polyether ether ketone
PLA	Polylactic acid
PP	Polypropylene
PBE	Propylene-based elastomer
RP	Rapid prototyping
SEBS	Styrene-(ethylene-butylene)-styrene
TPE	Thermoplastic elastomer
3D	Three-dimensional
TO	Tin octoate

1. Introduction

In this current era of technological advancement, three-dimensional (3D) printing has attracted increasing attention worldwide. It is also well known as additive manufacturing (AM) technology [1–8]. 3D printing is defined as AM since it requires adding materials together, which means layer by layer in sequence on an even plane to create actual structures of objects from a digital 3D layout [2,3,7,9–12]. In the 1980s, the fused deposition modelling (FDM) method evolved as a part of 3D printing technology [13,14]. This approach is one of the technological advances studied and developed by researchers and industries as part of rapid prototyping (RP) engineering [15,16]. A variety of sectors, like building, mock models, commodities production, medical, electrical, and engineering use FDM [2,5,17–24] because of its instantaneous production capacity with a lower cost [15,24–29].

The FDM technique is also known as fused filament fabrication (FFF) [30–33]. In terms of production, the FDM method plays a role character in injection molding [15,25]. The core idea behind the FDM fabrication technique is to melt the feed filament and mold it to create new forms [17,20,27,34,35]. The filament is wounded over a spindle, driven by a driving wheel to the nozzle head. Then, the filament is melted by applying heat to a molten state and then heated to semiliquid in a temperature-controlled nozzle head. The nozzle accurately extrudes and directs materials into thin layers like fibers to build structural components [3–5,7,8,11,36–39]. The computer-aided design (CAD) software installed on the computer controls the whole printing process [3,4,6,8,10,11].

FDM is popular and preferable compared to other AM methods. As the filament preparation is easier, FDM is highly adaptable for all kinds of industries and it is also affordable [16,36,40–43]. There are many different types of materials available in response to market demand for a variety of purposes [44–46]. FDM can generate a variety of complicated forms and structures [1,9,11,47,48] using 3D model data while effectively managing resources, resulting in reduced waste and a variety of additional benefits over existing production [13,15,17,49]. Furthermore, simulation is applicable for FDM, which allows the prediction of material's behaviour prior to the real 3D printing process. Based on the simulation, changes will be done to meet the requirements of 3D printed parts [45]. 3D printer is suitable to be used at all levels as it is user friendly due to the simple process in creating 3D printed parts [44,46] and fit for low volume of production [45,50]. Additionally, FDM printer is available in various sizes therefore, it does not consume huge space [46] and available in affordable price [50,51]. In the 3D printing industry, filament made from thermoplastic is widely used because thermoplastic has lower melting points [3,19,20,52–54]. The widely utilized filament materials are acrylonitrile butadiene styrene (ABS) [16,19,34,35,55–59], polylactic acid (PLA) [33,35,53,55,57], polyether ether ketone (PEEK) [60–62], and thermoplastic blend [34]. Thermoplastic has had a prominent application in the evolution of filaments since the advent of FDM.

However, existing filaments do have drawbacks. In line with this proposition, ABS is required to operate under higher temperatures and release toxic fumes which may harm the environment [33,63,64]. A warping issue resulting from high operating temperature will limit the adhesion mechanism to the printing platform and in between printed layers on the 3D printed part [52,65,66]. ABS filament also requires a printer bed with an enclosed environment to reduce the warping effect [52,67]. Besides, PEEK requires a higher operating temperature due to a higher melting point [68,69]. It is also highly crystalline and causes difficulties in the manufacturing process [51,62]. Additionally, ABS and PEEK are synthetic thermoplastic polymers made from crude oil as a source that has a longer shelf life and a slower deterioration rate, which eventually leads to environmental issues since it does not degrade in a short period [22,70,71]. To reduce warping and eliminate toxic emissions produced from 3D printing using ABS and PEEK, the implementation of new filament material is needed.

Despite PLA being a biodegradable material [6,8,12,17,38], it possesses similar mechanical properties as a synthetic polymer with lower processing temperature [72–74]. Moreover, the usage of PLA is preferred as printed parts are not easily warped and do not emit toxic fume or odor during the printing process [65,75–77]. Besides, the printing process using PLA does not need an enclosed environment, and a printer bed is not compulsory [52,73]. Despite all the above qualities of PLA filament, it is not suitable for high mechanical performance applications. The rigid structure of PLA contributes to

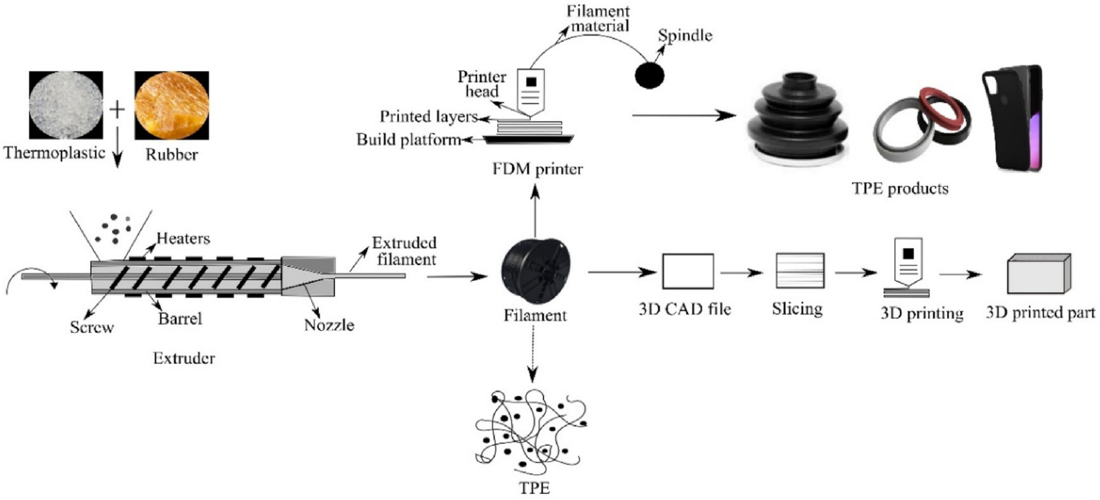

Fig. 1 – Filament fabrication and 3D printing with TPE material.

brittleness [78–80] and results in PLA's poor toughness and impact resistance [60,63,64]. Additionally, PLA resistance toward heat is lower due to its lower glass transition temperature (T_g) compared to ABS and PEEK filament materials. The processing temperature of PLA is only restricted to lower temperatures [81].

In correlation with that, to further enhance the qualities of PLA and suppress the restriction of PLA, elastomer material can be blended. Filaments developed using PLA thermoplastic and elastomers, which are known as thermoplastic elastomers (TPE) [74,82,83], can overcome the limitations of the current PLA filaments. PLA-based TPE filament materials exhibit improved properties and are a great substitute for FDM printers. As the TPE material is composed of an elastomer phase, the primary weakness of PLA, which is the brittleness will be overcome [74,77,84–89]. Therefore, the impact, toughness, and elasticity of PLA-based TPE will be higher than PLA filament alone. For the PLA-based TPE filament, mechanical characteristics, compatibility, and thermal properties are the three main considerations. These properties are dependent on the ratio, compatibilizer, and melt blend process [13,63,64,74,81–84,90–92].

However, research relating to the TPE filament fabrication from PLA and rubber remains limited. This review aims to offer a comprehensive summary of the filament materials, elastomer, and TPE. Besides, this review focuses on prior research studies on the characteristics of TPE that emphasize the material's ability for usage in 3D printers. The graphical illustration of filament fabrication and 3D printing with TPE material is presented in Fig. 1.

2. Filament material

FDM is popular because of its versatility of filament material. In the FDM printing method, a hot nozzle of the printer melts the filament fed into its melting temperature (T_m). Melted polymers become flexible and allow for the construction of desired forms on the printing platform before solidification. In the FDM technique, the product properties rely on the type of filament materials used. This part comprises the types of materials in 3D printing. Types of FDM filaments and characteristics are discussed in more detail in the next section.

2.1. *Synthetic thermoplastic*

ABS has also been widely applied in 3D printing since the beginning of evolution [56,59,93–95]. ABS is a synthetic polymer as it is a petroleum-based polymer that can be recycled but is not degradable. The presence of nitrile in ABS acts as an active site and provides polar properties for ABS, while styrene gives the product shiny properties as demonstrated in

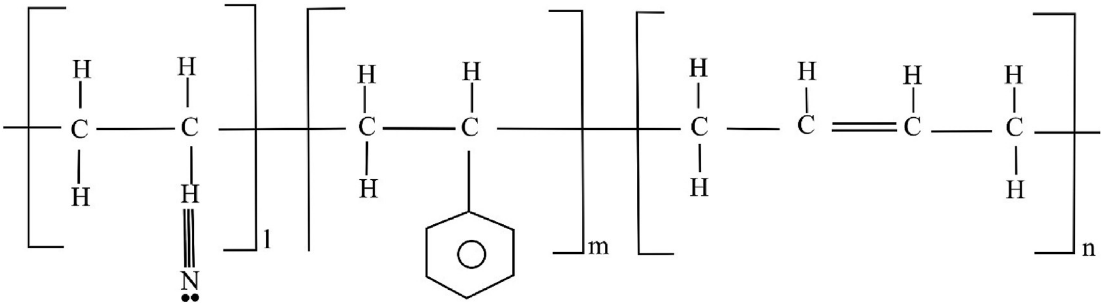

Fig. 2 – Chemical structure of ABS [96].

Fig. 2. Meanwhile, the butadiene makes ABS a material with better resilience [56,59,93–95]. Copolymerization between acrylonitrile and styrene provides strength and rigidity, while polybutadiene rubber (BR) imparts toughness in ABS, allowing it to be operated at a higher temperature during the extrusion process since it has high-temperature resistance due to acrylonitrile [66,94,97]. ABS is considered to be more economical as it is cheaper and exhibits excellent mechanical properties. It meets the qualities needed for engineering products in terms of capability to bear impact that causes damage [14,59,93,94].

ABS has excellent properties such as high toughness, rigidity, good thermal stability, and resistance to chemical and environmental attacks. It also has a low thermal expansion coefficient; hence, it is easy to mold products with good dimensional stability [59,94,98]. It also offers processing at a higher temperature, because it has a higher T_g. Therefore, it is thermally stable to withstand degradation even due to exterior application [14]. It is known that ABS has been used to toughen the brittleness of PLA, but the modulus and strength properties reduced significantly [98]. No crystallization has been observed in amorphous polymers like ABS, but it has a higher shrinking percentage on its own. In comparison, ABS has a larger temperature difference and tension during the FDM printout with higher melting and printing temperatures than PLA [99].

Abrasive and impact resistance make ABS an excellent thermoplastic for a wide range of applications. Its outstanding mechanical characteristics, thermal resilience, mild flexibility, and extended serviceability make this polymer a great choice for FDM production. In addition, the rheological property of ABS is shear thinning. Therefore, the viscosity reduces during processing as heat is involved and increases the flow ability during printing [94]. One of the issues in 3D printing with ABS material is warping. The cause of these defects is internal thermal stress and unequal cooling of layers of the 3D model which result in a curve [52,66]. Due to its tendency to shrink and bubbles on the finished surface [79,100], the fabricated layer does not adhere properly and causes delamination [33,52,101]. Besides, when ABS filament is operated at higher printing temperatures (210–260 °C), toxic fumes are emitted and it is not environmentally friendly [33,63]. Apart from that, ABS has lower dimensional accuracy and it is highly influenced by variation of the nozzle temperature [57].

Additionally, in 3D printing, PEEK can substitute as a filament especially in the medical field as it is biocompatible due to its non-toxic properties [78,102], its high melting point (>350 °C), and small range of optimum temperature (360–400 °C) [68,79] for printing. PEEK is composed of aromatic groups, ether, and ketone groups as illustrated in Fig. 3. The T_g of PEEK lies between the range of 143–148 °C which provides exceptional thermal characteristics [68], the capability to operate at higher temperatures [103], and resistance to high thermal energy [35]. Moreover, PEEK offers better mechanical properties due to its high molecular weight [68,103,104] with a raster angle of 45°/45° [68]. PEEK is referred to as an engineering plastic because it has excellent characteristics in a semi-crystalline state [35,68]. Apart from that, PEEK has outstanding abrasion resistance, durability, and resistivity toward chemicals. Moreover, PEEK is also not easily hydrolyzed with water as a solvent [103].

However, the usage of PEEK is not wide due to its major disadvantages where it is expensive relative to other thermoplastics. Furthermore, its semi-crystalline structure causes complexity during manufacturing [68,103]. Besides, the utilization of PEEK for FDM printers is a challenging material due to the necessity of higher printing temperatures [102]. Due to its higher T_m, a higher temperature within a range of 380–410 °C is required [105]. PEEK tends to cause damage to the printed parts due to thermal stress, which is heat unequally transferred between layers. As a result, thermal fractures will develop on the surface, leading to poor surface finishing [102].

2.2. *Biodegradable thermoplastic*

In the current 3D printing industry, the use of a few filaments made of biodegradable materials, such as PLA and polycaprolactone (PCL) [82,106], and polybutylene adipate terephthalate (PBAT) is increasing [107–109]. PCL is less rigid and has lower thermal stability compared to PLA due to lower T_g at (60 °C) [110–112]. It also has a lower processing temperature than other conventional filaments like ABS, PEEK, and PLA due to its low T_m [82,110] and is widely used for medical applications [113,114]. The drawbacks of PCL are its highly flexible nature and poor mechanical properties [112,115,116].

On the other hand, PBAT is a kind of thermoplastic elastomer copolymer. It behaves more like an elastomer with higher ductile properties [117,118]. There PBAT exhibits excellent toughness and is extremely flexible with a higher elongating ability [109,119]. However, the application of PBAT is limited in 3D printing applications due to its higher price and very low rigidity with lower strength [118,120,121]. Due to the above limitations of PCL and PBAT, the usage of PLA filament is evolving at a faster rate and has become an excellent choice of material in the 3D printing industry to overcome the existing drawbacks of ABS and PEEK filament [8,12,71,72,84,122].

PLA as illustrated in Fig. 4 is a well-known polymer in FDM technology as it is an essential biodegradable polymer with excellent mechanical properties [6,12,17,38]. Through the carbohydrate fermentation obtained from crop products such as maize, PLA is created using lactic acid [6,84,85,89,124–127]. Since its production involves abundantly found sources, the

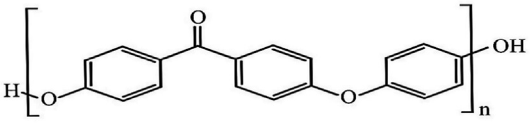

Fig. 3 – Chemical structure of PEEK [53].

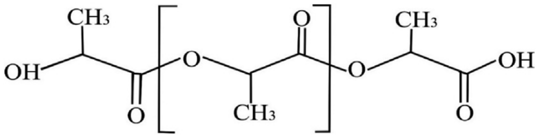

Fig. 4 – Chemical structure of PLA [123].

cost of PLA is relatively cheaper [13,85,126,128]. Among researchers, PLA is the most familiar because of its multiple characteristics resembling the current synthetic polymer [80,129]. PLA is a group of aliphatic polyesters that can be produced using alpha–hydroxy acids [86,92,130,131].

Additionally, PLA has excellent mechanical properties where high modulus, strength, and superior transparency are equivalent to traditional polymers [4,37,71,78,79,85,124,132]. PLA has a low melting point with 60–65 °C T_g [71,127,133,134]. The bonding through fusion between layers of PLA takes place at 155–185 °C [127]. Despite composability, PLA is less preferable as a substitute in broad applications of commodities polymers [78–80] due to some restrictions. PLA has several disadvantages, including its extreme stiffness, poor impact resistance, ductility, low thermal resistance, and low elongation at break. It can withstand brittleness [71,80,89,124,132,135–137], has relatively slow crystallization, and has gas permeability [78,79,85,125]. PLA is also categorized as having Newtonian behavior that causes difficulties to control the viscosity during processing [138] due to low melt flow index (MFI) [80]. As the temperature changes from lower to higher during processing, the mobility of PLA molecules increases [139].

To modify the thermal and physical characteristics of PLA, it is needed to be blended with some other polymers and additives [71,126,128,130,135]. Several researchers have stated in the past that incorporating it with a variety of flexible polymers that could work as toughening agents has increased its strength [84–89] such as elastomers with PLA, notably degradable natural rubber (NR) [89,124,140–142]. This method eliminates this shortcoming in the mechanical characteristics of PLA [137,143]. Blending PLA with rubber is a more economical method and convenient to improve certain qualities [86,89,92,125,130]. Apart from that, the inclusion of modifiers will cause changes during the reactive blending [33,137].

Both ABS and PEEK must be printed at a very high temperature and leading to warping issues. Additionally, both materials are derived from petroleum, they cannot degrade. As a result, this is not ecologically friendly despite having great temperature stability. Therefore, the warping issues and toxic fume emissions can be avoided by using PLA, which is biodegradable. Besides, PLA is printed at lower printing temperatures within a range of 180–230 (°C) [105,144] due to its lower melting point. PLA can also be printed at lower temperatures without implementing a heated bed which could also reduce warping issues [76,144]. However, when subjected to a lot of stress, 3D parts created from PLA will deform easily due to their brittleness. The addition of elastomer into PLA will reduce the deformation by suppressing the brittle behavior. An elastomer is a tough elastic material that could deform and is capable to return to its original form when the subjected force is eliminated. The rubber particles dispersed in PLA will act as stress absorbers. Under stress, a thermoplastic elastomer blend will form cracks rather than completely break. It indicates that rubber acts as a stress concentrator. Through these cracks, stress is released instantly without severe damage in TPE [145]. The general properties of filament materials used in 3D printing are presented in Table 1.

Table 1 – Comparison between all filament material.

Material	Glass transition temperature (°C)	Melting temperature (°C)	Printing temperature (°C)	Thermal resistance	Degradability	Application
ABS	105–115 [61]	No true melting points [146,147]	210–260 [105,144]	Excellent [14,66,97]	Non-Biodegradable [70,95]	Engineering products [14]
PEEK	143–148 [68]	330–340 [68]	380–410 [105]	Excellent [35,68,103]	Non-Biodegradable [68]	Biomedical [78,102]
PLA	60–65 [127]	155–185 [127,128]	180–230 [105,144]	Poor [135–137]	Biodegradable [84,85,125]	Commodity products [126,127]

3. Elastomer

NR and epoxidized natural rubber (ENR) belong to the elastomer category and are also considered biodegradable polymers. Furthermore, they possess a lot of excellent properties in terms of mechanical properties, heat resistance, abrasion, and resilience. Besides, elastomer also demonstrates higher elasticity [84,125,148]. ENR is a cis-1,4 polyisoprene [84,85,90] which is an NR modification [85,125]. NR is chemically modified with acetic acid or formic peroxy acid, and the in–situ reaction is involved in the synthesis of ENR, indicating that NR can be easily manipulated with a polar monomer via grafting or epoxidizing [96,99,100]. The epoxy group formed into NR via in situ epoxidation will act as an active site for crosslinking and providing polar properties [85–88,125] with the presence of an epoxy ring and contain a double bond that reacts by undergoing crosslinking reaction [135]. The T_g of ENR is −21 °C [125] while the T_g of NR is −70 °C [149]. A shift in T_g is noted, where T_g of ENR-PLA is higher when the ENR is copolymerized with PLA [125].

Additionally, the ENR is used in the blending process in the second phase and acts as a toughening agent [84,85,90,125,135] without losing its characteristics [150]. ENR induces cold crystallization mechanism in PLA and acts as a nucleating agent to improve toughness. The fracture energy is absorbed by second phase rubber because rubber performs a role as a stress concentrator [85,135]. The microvoids produced by the rubber particles act as craze starters and inhibitors. The high number of voids tends to create a large number of crazes to provide a yield of shear by absorbing the impact energy [78,79,85]. Besides, cost-effectiveness, renewability, hardness, and durability are advantages of ENR [78,79,151]. Moreover, ENR offers higher impact properties and, resistance toward tear property is high [85,135]. Besides, the elastomers like NR and ENR will undergo shear thinning behavior. As ENR is more polar than NR, it possesses slightly higher viscosity and shear thinning. Mastication of elastomer will reduce the viscosity [152]. Due to the numerous advantages of the elastomer phase, it is widely incorporated into thermoplastic as a blend material for TPE. However, blending of PLA/elastomer will produce an incompatible blend due to the polarity differences. The difference in polarity acts as a hindrance in obtaining [87,88]. The inclusion of a compatibilizer will effectively promote and improve the compatibility between PLA/elastomer. Therefore, for further enhancement of the miscibility, effective compatibilizer incorporation is necessary which is discussed in the following section.

4. Compatibilization

Compatibilization is a mechanism that promotes homogeneity in an immiscible polymer blend. Compatibilization promotes reduction in interfacial tension which eventually reduces the phase separation that exists in an immiscible polymer blend. As a result of effective compatibilization mechanism, interfacial adhesion will develop and stabilize the blend [153,154]. There are two types of compatibilization mechanisms for immiscible polymer blends.

The first category of compatibilization is by adding a third component called compatibilizer to an immiscible polymer blend. It is also known as cosolvent where the compatibilizer interacts with both phases and the compatibilizer is usually block copolymer or graft copolymer [153,155]. The segments of copolymer compatibilizer will aid in dispersion of isolated phases in immiscible blend and allow stabilization of phase morphology [156]. With more homogenous dispersion, stronger adhesion between phases [151] will be formed. It is significant in altering the properties of the blend by decreasing the interfacial tension across the interface of polymers involved in a polymer blend system [154].

The second one is chemical modification of polymer interface via chemical reaction and it is known as reactive compatibilization [153–155]. The multiple functional sites of the reactive compatibilizer have a higher tendency to react with the polymer's functional group. It is highly suitable to be used in the melt blending technique [157,158]. A reactive compatibilizer is commonly used for compatibilization due to its high reactivity rate, shorter reactive duration, lower viscosity, and affordable price [157–160]. Besides, in situ reaction is more preferable compared to chemical treatment because of the absence of solvents that are harmful to the environment [161]. Reactive compatibilizer contains reactive sites such as epoxy, anhydride, and isocyanate. In situ reaction takes place in reactive compatibilization and results in the formation of copolymers that are arranged into block and graft copolymers [155,159,160,162].

5. Thermoplastic elastomer (TPE) blend

TPE, which is also one of the polymer mixtures, is composed of an elastomer phase and a thermoplastic segment. TPE displays elastomeric characteristics at ambient temperature and can be reprocessed and recycled as a standard thermoplastic material [163–165]. The rigid segment imparts glassy blocks of thermoplastic with high T_m and T_g. The soft part reveals rubbery blocks with low T_g [165]. TPE possesses pseudo-plasticity nature, hence, can utilize the same process as a thermoplastic polymer [166].

TPEs are used in various industries due to TPE's excellent mechanical characteristics. Furthermore, TPE exhibits both toughness and elastic capability. TPE also enhances material rigidity and resilience via integrating transitory dynamic bonds [167,168]. Another attractive characteristic of TPEs is low density and satisfactory thermal stability. TPE also has a better surface leakage tolerance, the ability to maintain a good hydrophilic nature, the ability to resist ultraviolet, a wide temperature range, and a low cost of output [165]. TPEs are safer for our environment as it is recyclable and does not involve crosslinking agent [163–165]. Scrap and waste are inevitable in any manufacturing process, and TPE waste is reusable because it does not undergo the cross-linking process [163]. With the above qualities, the TPE blend is considered a good filament material for FDM. Over the years, many researchers have performed experimental work by incorporating NR or ENR into PLA to enhance the properties of PLA which will be reviewed in the following sections [123,130,132,135,136].

6. PLA/rubber TPE blend

The ability to overcome the problems of PLA thermoplastic filament will be enabled by combining PLA and rubber such as NR and ENR. To begin, it will address the issue of poor toughness and brittleness of PLA despite having higher mechanical properties in the aspect of tensile strength and modulus. Apart from that, the presence of elastomeric material will create a material with elastic behavior and improve the toughness of PLA. TPE blending of PLA and rubber is recyclable and safer for the environment as both PLA and elastomer are derived from natural sources. Many researchers studied the PLA/NR and PLA/ENR blends to determine the blend properties [84,85,88,135,137,140,141,148,152,169,170].

6.1. *PLA/NR*

The summary of studies related to PLA/NR and compatibilizer used for PLA/NR are listed in Table 3. The impact of epoxidized soy oil (ESO) as a compatibilizer in the PLA/NR mixture was studied by Burkov et al. [137]. The inclusion of ESO enhanced the poor compatibility of the blend. The compatibilizer enhanced the thermal stability of PLA and promoted flexibility. Meanwhile, the size of NR regions reduces to allow more homogeneity while the mobility of PLA increases.

Similarly, Udomkitpanya & Srikulkit [140], also studied the compatibility of the PLA/NR blend by adding NR-g-Poly (acrylic acid) (PAA) as a compatibilizer. The finding showed that the inclusion of NR-g-PAA considerably enhanced the elongation, impact strength, and heat resistance of blends. 30 wt% compatibilizer was found to be the most effective to increase PLA's mechanical characteristics.

Besides, Sookprasert & Hinchiranan [141], evaluated the PLA/NR blend with different amounts of NR-g-PLA compatibilizer (1–5%). The study found that the longer the grafting reaction period, the higher the number of grafted copolymers formed. A similar trend was observed for the rise in reaction temperature. The inclusion of a higher graft compatibilizer into the blend gives a positive impact by enhancing the impact strength and reducing the phase separation.

Additionally, Xia et al. [170] explored the impact of glycidyl methacrylate (GMA) compatibilizer grafting with NR. The increase in GMA compatibilizer and dicumyl peroxide (DCP) initiator concentration has a beneficial effect on grafting yield, the proportion of gel, and Mooney viscosity. These data show that the grafting reaction continued to grow with the concentrations of the GMA and DCP as it will lead to increased formation of the interlinked interaction.

Following that, Wongngam & Pattamaprom [148], examined the effect of tin octoate (TO) as a catalyst in the grafting reaction of NR with maleic anhydride (MA), NR-g-MA in the PLA/NR blend. The grafting reaction of NR-g-MA with and without the catalyst was found to be similar. With TO catalyst for the grafting process, however, less incomplete grafting residue was produced and higher grafted copolymer PLA-NR was yielded.

For PLA/NR most researchers used graft compatibilizer as stated in Table 2 [137,140,141,148,170]. The copolymer is a graft compatibilizer and synthesized through the in–situ reaction. It usually involves two different functional groups and monomers to build a network through grafting. Graft compatibilizers reduce the tension that existed in the TPE blend better than homo-compatibilizer [87,125]. The graft compatibilizer contains functional groups of both phases involved and it will interact with the interface of the polymer matrix during melt blending [81,125]. A better adhesion force will develop and strengthen the polymer blends' mechanical properties by inducing better interactions. The incorporation of a compatibilizer will promote homogeneity by eliminating phase isolation in the TPE blend. Overall, the inclusion of a compatibilizer reduced the T_g and T_m of the two-phase blend, resulting in improved thermal stability.

6.2. *PLA/ENR*

The summary of studies related to PLA/NR and compatibilizer used for PLA/ENR are listed in Table 3. Syed et al. [84] investigated the influence of liquid epoxidized natural rubber (LENR) as a toughening agent in the PLA system. The impact property of PLA improved with LENR as a toughening agent.

Table 2 – A summary of ratio and compatibilizer used for PLA/NR.

No	Researchers	Blend	Ratio	Compatibilizer	Findings
1	Burkov et al. [137]	PLA/NR	60/40	ESO (5, 10, 15, 20 pph)	• ESO improves compatibility, thermal stability, and flexibility. • The blend becomes homogenous with the reduction of NR regions • The mobility of the PLA increased
2	Udomkitpanya and Srikulkit [140]	PLA/NR	100/0, 90/10, 80/20, 70/30	NR-g-PAA (0, 10, 20, 30 wt%)	• NR-g-PAA improves the elongation, impact strength, and thermal properties
3	Sookprasert and Hinchiranan [141]	PLA/NR	80/20	PLA-g-NR (1–5% (w/w))	• Longer grafting reaction time and high temperature increase the formation of grafted copolymers. • Graft copolymer improves impact strength and reduces phase separation.
4	Xia et al. [148]	PLA/NR	–	NR-g-MA	• A rise in GMA compatibilizer and DCP initiator increases the grafting yield. • Enhanced the interlinked interaction
5	Wongngam and Pattamaprom [170]	PLA/NR	70/30	NR-g-MA	• The presence of catalyst promotes higher grafting

Table 3 – A summary of the ratio and compatibilizer used for PLA/ENR.

No	Researcher	Blend	Ratio	Compatibilizer	Findings
1	Syed et al. [84]	PLA/LENR	100/0, 95/5, 90/10, 85/15, 80/20	LENR	• LENR enhances the toughness and increases the impact of energy • with 10 wt% of LENR homogenous dispersion is achieved. • Tensile properties dropped with LENR except for elongation. • Higher LENR concentration decreases the T_g and T_m.
2	Abdullah and Mohamad [135]	PLA/ENR	90/10, 80/20, 70/30	–	• TS and the modulus decrease with ENR. • Crosslinking agent increases the bonding features and rigidity.
3	Abdullah et al. [85]	PLA/ENR, Graphene loading (0.5–2.0 phr)	90/10, 80/20, 70/30	–	• With 10 wt% of ENR in PLA, a homogenous dispersion is achieved • ENR promotes stress transmission across the PLA and improves toughness • A higher ENR ratio reduced the thermal stability
4	Klinkajorn and Tanrattanakul [88]	PLA/ENR	90/10	ENR-g-MA (0.15, 0.30, 0.45,0.60 phr)	• A rise in MA concentrations increases the grafting reactions and compatibility. • The impact and thermal characteristics improved. • T_g of PLA reduced while T_g of ENR increased.
5	Snowdon et al. [152]	PTT/NR, Poly (tri – methylene terephthalate) (PTT)/ENR	0/40	DCP and Maleate BR (0.75phr)	• ENR offers lower interfacial tension with better adherence than NR because ENR is more miscible • Compatibilizers reduce the rubber domain size • DCP compatibilizer hindered the impact property. • Maleate BR has higher interaction with MA
6	Zhang et al. [169]	PLA/ENR	100/0, 95/5, 80/20	–	• The PLA/ENR blend is classified as amorphous, as crosslinking occurs between PLA and ENR when crystallization is inhibited. • The addition of ENR increased the PLA's thermal properties, impact resistance, and elongation significantly.

Table 4 – Types of PLA-based TPE filament used for FDM printer.

No	Researchers	TPE filament	Ratio	Filament diameter (mm)	Printing temperature (°C)	Bed temperature (°C)
1	Intan et al. [74]	PLA/NR and PLA/ENR-25	80/20	1.75	210	55–60
2	Fekete et al. [171]	PLA/NR	95/5 90/10 85/15 80/20	1.75	215	60
3	Arrigo et al. [172]	PLA/elastomer (Talc and calcium carbonate filler)	–	1.75	200	65
4	Fekete et al. [173]	PLA/NR	90/10	1.75	215	60

With 10 wt% of LENR, the blend obtained a homogenous dispersion that resulted in maximum impact property. Tensile properties, T_g, and T_m somewhat dropped as LENR concentration increased except for elongation with higher dispersion. Besides, Abdullah & Mohamad [135] mentioned that as the quantity of ENR rises, the TS and the modulus in PLA/ENR/ENR-TPV mixtures decrease. However, the vulcanized PLA/ENR mixed with the crosslinking agent increases the bonding features and provides rigidity. The length increases to 20 wt% more rubber. The PLA/ENR mixtures were co-continuous, whereas the PLA/ENR-TPV mixtures exhibited a spread of ENR rubber particles.

Furthermore, Abdullah et al. [85] studied PLA/ENR with different loadings of ENR and graphene. With 10 wt% of the ENR loading, the PLA/ENR blend reaches a finer dispersion, and stress transmission increased with excellent infraction across the PLA and ENR phases. With higher loading of rubber, the impact strength increases, but the thermal stability drops. In addition, Klinkajorn and Tanrattanakul [88], found that graft copolymer production increases with increasing MA concentrations in grafting reactions, while compatibility improved linearly. Hence, the impact characteristics and thermal stability of both PLA and ENR improved where the T_g of PLA dropped, while the T_g of ENR increased.

Additionally, Snowdon et al. [152] produced two distinct NR and ENR rubber blends using poly (trimethylene terephthalate) (PTT). The effects of DCP and maleate BR were investigated as compatibilizers. ENR mixture offers better adherence and miscibility than NR. However, the DCP hindered the impact property. Meanwhile, the maleate BR has higher interaction with MA which strengthens the impact property and provides yield elongation. In another study, Zhang et al. [169] investigated the properties of the TPE blend made from PLA/ENR. From the data obtained, the studies found that the TPE blend has a shear-thinning rheological behavior as a result of interaction across the functional group of polymers involved in a blend. The blend's complex viscosity is higher than neat PLA. A higher viscosity will lead to poor flowability. However, it also enables compactness in the extruded filament.

The findings highlighted that the mechanical properties improved significantly in the PLA/ENR blend. Apart from that, the ENR phase into PLA improves the impact characteristics, particularly strength and toughness. Therefore, the TPE blend allows efficient transfer of stress. In addition, ENR enhances thermal stability by lowering the T_g and T_m as the blend phases become compatible. Compared to NR, ENR is more compatible with PLA as ENR acts as a compatibilizer and a high adhesion force is developed. Based on the findings on the properties of PLA/NR and PLA/ENR, it is possible to prepare TPE filament using PLA and NR or ENR. So far, however, only a small number of researchers have examined PLA/rubber as a filament for FDM. In the next section, experimental works on PLA-based TPE filaments and some other thermoplastic-based TPE filaments are discussed.

Table 5 – Types of non-PLA-based TPE filament used for FDM printer.

No	Researchers	TPE filament	Ratio	Filament diameter (mm)	Printing temperature (°C)	Bed temperature (°C)
1	Sayanjali et al. [19]	ABS/SEBS	90/10 80/20 70/30 60/40	1.75	235	105
2	Banerjee et al. [83]	PP/SEBS	40/60	2.80	220	100
3	Wissamitanan et al. [82]	PCL/NR and PCL/ENR-50	50/50	1.75	–	–
4	Siqueiros et al. [174]	ABS/SEBS	100/0 75/25 50/50 25/75	1.75	230–265	110
5	Eutionnat-Diffo et al. [175]	LDPE/PBE	100/0 80/20 60/40	1.75	245	–

7. TPE filament

In FDM manufacturing, the usage of TPE filament is still new. It appears that a lack of related research studies exists for PLA-based TPE filament material for FDM. Therefore, in this section, apart from PLA/rubber filament, TPE filaments made from different thermoplastics are also reviewed such as polypropylene (PP), PCL, and low-density polyethylene (LDPE). The types of PLA-based TPE filaments used in recent studies are listed in Table 4. Research work related to non-PLA-based TPE filaments is tabulated in Table 5.

Intan et al. [74] experimentally examined TPE filament for FDM printers made from PLA/NR and PLA/ENR. TPE filaments are prepared in different ratios to improve its physical properties and mechanical properties. Rubber addition has reduced the tensile strength of PLA and, on the contrary, the elongation denoting flexibility has improved. An 80/20 ratio of PLA/ENR is the optimum ratio to enhance the mechanical characteristics of the 3D printed parts. According to the morphological analysis, the size of rubber is increasing along with the addition of rubber. The size of the NR particle was bigger than ENR. The larger size of rubber particles will act as stress concentrating regions to avoid the deformation of PLA regions. PLA's thermal characteristics were dramatically changed with the addition of NR and ENR-25 where the T_m reduced. Drop-in T_m reveals the nucleating effect of rubber particles. The higher the melt index, the better the printability. The PLA/ENR-25 has a higher printing ability than PLA/NR. With a fixed bed temperature in the range of 55–60 °C which is closer to the T_g of PLA, both PLA blends can be manufactured satisfactorily. The ENR-25 rubber blend with PLA becomes tacky during the printing process. Under a fixed bed temperature, both PLA blends can be manufactured satisfactorily. An ENR-25 modifier can cause tackiness during the printing process.

According to studies conducted by Fekete et al. [171] on PLA-based TPE filament using NR as elastomer phase, it was identified as suitable for 3D printing. The researcher has prepared filament filled with various amounts of elastomer loading with a range of 0 to 20 wt%. The experiment found that the inclusion of rubber resulted in poor bonding between the layers due to weak force of adhesion which was identified by morphological studies. Despite the weak bonding, the toughness of the filament and elongation behavior had improved along with NR content. Meanwhile, the tensile strength and modulus dropped as the rubber domain introduced elastic behavior that suppresses the strength of PLA to withstand the stress and prevent deformation. The 3D printed part with loading of 20 wt% of NR exhibits the highest mechanical properties as the force acting was absorbed by rubber particles.

For the thermal behavior, the addition of NR has no influence on the Tg while the PLA/NR TPE filament exhibits two Tm peaks representing various PLA regions. The study highlighted a significant finding where the rubber size particle incorporated into the PLA increases along with the ratio. The average size of rubber particles was 0.36 μm for 5 wt.% of NR and 1.64 μm for 20 wt% of NR. A rise in size relates to the occurrence co-continuous phase and results in increasing rubber domain size. The researcher concluded that 20 wt% of rubber has reached higher dispersion and resulted in a homogenous TPE blend. The larger the rubber phase, the higher the elongation of filament with lower tensile strength and modulus with flexible nature. As written by Zhao et al. [130], toughening effect and improvement in mechanical properties of PLA blend system depend on the size of rubber particles. Rubber particle size was identified as an important element in toughening mechanism since enhancement of PLA/elastomer system relies on homogenous dispersion of toughening agent which are the rubber particles. Greater surface area is available for stress energy transmission when rubber particles are smaller. However, extremely small rubber particles will lead to a weak crazing mechanism, which leads to failure in reducing stress. On the other hand, larger particle size of rubber also suppresses the ability of crazing due to formation of coalescence. In accordance with this, optimum rubber particle size is necessary to promote the role of rubber as stress energy absorber for PLA blend system for maximum enhancement.

Arrigo et al. [172] studied the printability of PLA-based filament of various types, including TPE filament. The PLA-based TPE filament was fabricated using elastomer and mineral filler such as talc and calcium carbonate. The TPE filament exhibits non-Newtonian behavior and a higher shear-thinning rate at a higher frequency. This kind of slow behavior is beneficial for the processing of the blend via extrusion and ensures that the extrudate maintains its configuration during the extrusion process. Additionally, shear-thinning flowability avoids the occurrence of filament buckling and shows good printing ability.

Besides, Fekete et al. [173] conducted studies on PLA-based TPE filament using NR. The effect of rubber loading on the different types of PLA was analyzed. Rubber was included in PLA as a toughening agent in TPE filament fabrication. Due to the rubbery nature of NR, all kinds of PLA based TPE 3D printed parts experienced a reduction in tensile strength and tensile modulus regardless of the infill pattern applied during printing. On the contrary, the elongation of filament has improved for all types of PLA. The researcher also highlighted that a higher toughening effect was obtained by including rubber in PLA, especially when printed under a parallel direction with higher interfacial adhesin across the printed layers formed.

According to Table 2, the blending of PLA and NR or ENR is printable and demonstrated improved properties in the aspect of elongation, impact properties, and thermal properties compared to PLA filament. The flow behaviour of the PLA/rubber blend also exhibits shear thinning behaviour with non-Newtonian rheological property, which is favourable for 3D printing. The printed layers exhibit good structural stability, thus the 3D printed will have high dimensional accuracy. The printing temperature and bed temperature applied are lower. On the other hand, based on the previous study reviewed in subsection 6.2, it has been found that inclusion of nanofiller filler has been done for PLA/ENR TPE blends. However, it seems that no research has been done to determine the impact of filler incorporation on the TPE filament because studies that are specifically related to the filler added to PLA-based TPE filament are rarely found. This review paper did not concentrate on nanofiller incorporated TPE based filaments due to the relatively limited amount of prior research.

Sayanjali et al. [19] explored the use of ABS and styrene-ethylene/butylene-styrene (SEBS) to make TPE filament for 3D printing by varying the ratio of SEBS to ABS blend in the filament. When SEBS content in the ABS/SEBS blend was increased, the MFI dropped. After the addition of the elastomer phase to thermoplastic, the blend experienced a shear-thinning behavior due to the nature of the elastomer. The printability of the blend improved as the shear rate and melt flow resistance during the printing process reduced. The flowability of the material via the nozzle reveals its printing capability. Because of the shear-thinning behavior, the TPE blend prevents clogs at the nozzle. On the other hand, due to shear-thinning rheological properties, the extruded filament will adhere to the others with good interaction. In the presence of elastomer material with elastic properties, the die swell of filament extruded increases. The die swell allows the filaments to be deposited and compacted more effectively. As a result, the gap between the extruded filament decreases, and the interfaces between neighboring filaments increase, which eventually forms stronger adhesion. There were fewer cavities across layers in printed samples when SEBS was incorporated. With the inclusion of SEBS, TS was marginally lowered and elongation at break was enhanced. In addition, the anisotropic in the mechanical characteristics of FDM components was decreased in blends. Therefore, when SEBS concentration in blends increases, its rheological characteristics resemble those of SEBS.

Following that, Banerjee et al. [83] prepared printable TPE filament with PP/SEBS. The blend filament was prepared with a twin-screw extruder within a temperature range of (190–200 °C) and an extruder speed of 190 rpm. The researchers performed a printing test with an FDM printer by using the adhesive sheet as a prevention method for the warping effect. A ratio of 20/80 is not printable as the filament was stuck in the nozzle, while 40/60 of PP/SEBS is considered suitable as a higher content of elastomer increases the elasticity properties with higher complex viscosity. The complex viscosity of SEBS is higher than PP. However, SEBS is classified as having shear thinning behavior.

Following that, Siqueiros et al. [174] studied a new TPE filament material made from ABS/maleated SEBS blend. A substantial shift in filament's mechanical properties was discovered, where compatibilized elastomer inclusion into thermoplastic produces elastic properties. The pure ABS has lower MFI (7.42 g/10 min), which indicates higher viscosity compared to ABS/maleated SEBS between 15.35 and 28.34 g/10 min. Higher MFI provides better printability. By blending a material commonly used in 3D printing ABS with an elastomer to modify its elastic behavior, a drastic difference in mechanical properties and failure characteristics was observed. In terms of material printability, we have determined that the MFI range for a rubberized thermoplastic material can be between 15.35 and 28.34 g/10 min.

On the other hand, Eutionnat-Diffo et al. [175] also created TPE filament with LDPE/propylene-based elastomer (LDPE/PBE). The addition of elastomer leads to an increment in MFI of the TPE blend up to 18 g/10 min with 60% of PBE, which eventually improves the ability of filament material to flow with lower resistance during deposition into layers. Furthermore, Wissamitanan et al. [82] tested TPE filament for 3D printing. Flexible TPE from PCL blend with NR and ENR was prepared with multiple ratios. The TPE material as filament for FDM was studied by analyzing the T_m and melt flowability. Owing to its high compatibility, homogeneity, and flow, the PCL/ENR blend outperformed PCL/NR for printing applications with better MFI. With a ratio of 50/50 PCL/ENR, tidy printable filaments with even diameters were produced.

Based on the finding above, non-PLA-based TPE filament produced using PCL, PP, ABS, and LDPE requires a higher temperature for printer bed and printing. The setting for printer beds with a temperature range of 100–110 °C and the range of printing temperature lies between 210 and 235 °C [19,48,74,82,83] was applied for non-PLA-based TPE material. Regardless of the type of thermoplastic and rubber, all the above TPE filaments have good printing ability and impact properties.

In the engineering field, the filament materials are chosen according to the end product's functionality since the properties of the filament materials affect the capability of functional part to perform its purpose as designed. Besides, profit and quality of 3D printed part are always considered as the primary factors for commercialization. Thus, for selection of filament materials needs to consider external factors like functional part's environment to ensure that the properties of 3D printed do not deteriorate. For higher profitability, an engineer is always required to look out for a filament material that is able to serve the purpose at lower cost. Despite the ability of TPE blend to achieve good 3D printed parts, 3D printing using TPE filament will face some difficulties. The problems and solutions for 3D printing TPE filament are reviewed in the following section.

8. Problems of 3D printing using TPE filament

In recent times, various types of TPE filaments are available in the 3D printing industry. Based on several studies using TPE filament, it was noticed that few potential problems arise during the 3D printing of TPE filament such as in a study by Kasmi et al. [176] who studied PLA/TPE filament for FDM, and found that the printed specimen had a higher porosity. The printed specimen contained higher porosity when printed with lower bed temperature because the printed layers cooled rapidly and thermal fusion across the layers is less. Therefore, there were numerous interfaces of printed layers that had many open fillings. Elevating the bed temperature allows for better heat diffusion and improved mechanical performance.

By reviewing a research conducted by León-Calero et al. [177], retraction of TPE filament during printing is difficult due to its flexible nature which leads to stretching filament material. Retracting filament movement is important to prevent filament from being printed into small strings. Printing with a lower speed helps to prevent printing TPE layers into strings and eventually avoids blockage of the printer nozzle. In a research conducted by Nakajima et al. [178], printing with TPE filament under higher printing speed caused nozzle clogging resulting in poorly printed parts. Under higher speed, the TPE filament material is not completely melted and tends to get stuck. However, excessive low printing speed resulted in drizzling of molten material and an uneven surface. Therefore, printing with optimum speed will avoid the difficulties in handling TPE filament during 3D printing. This finding is similar to the finding by Kalman et al. [179] where this experimental work also experienced jamming of TPE filament inside the nozzle. This problem was handled by reducing the distance between filament feeding and nozzle.

Narendra et al. [180] reported that TPE filament usually has low compressive strength than rigid material. Therefore, during the feeding of the TPE filament to the nozzle, the filament will bend out and interrupt the deposition of material through the nozzle. This phenomenon is known as the buckling effect and the high melt viscosity of TPE filament is one of the causes. To prevent buckling, these researchers suggested the usage of a material deposition tool where the TPE feeds in pellet form to be printed out directly. As stated by Kalman et al. [179], buckling can be avoided by reducing the printing speed. As per the findings of Fekete et al. [171], TPE filaments exhibit poor interfacial adhesion compared to PLA thermoplastic alone. This phenomenon is related to the presence of rubber in the TPE filament, where the rubber particles resulted in rougher printed layers. As the printed layers are not smooth, the adhesion formation was affected by the development of voids.

Based on the problems encountered during 3D printing of TPE filaments, proper handling of filament and optimization of printing parameters can overcome all the difficulties. By applying the right troubleshooting method, the above problems like buckling, jammed nozzle, poor adhesion, and porosity can be prevented, and obtaining a high-quality 3D printed from TPE filament is possible.

9. Summary and future works

This article provides a review of TPE material to show the potential of PLA-based TPE material for FDM to enhance the qualities of current PLA filament. There are TPE filaments in the market but the approach of manufacturing polylactic acid-based thermoplastic elastomer will be a new filament which is the novelty of this paper.

From reviewing the previous works, it has been concluded that producing TPE filament using PLA and rubbers is possible and printable with the right ratio, and applicable for 3D printers. The addition of rubber particles in PLA material can solve issues that traditional PLA thermoplastic filaments could not. In conclusion, the PLA-based TPE material exhibits outstanding mechanical and thermal properties making it appropriate for the FDM printer. Along with that, TPE must have compatibility characteristics to produce a miscible blend. For future proposal, research works need to be done to identify the processing parameters of TPE filament using the extrusion method and determine suitable parameter settings for FDM to print PLA-based TPE filament. A good quality filament prepared under optimum extrusion and printing with an optimized setting of FDM will deliver a 3D printed part with improved properties in various aspects namely mechanical, thermal, and structural stability.

In this study, several future works are suggested based on the identified gaps, as follows:

- Future research work needs to study the effect of the rubber particle size on the PLA-based TPE filament and analyse the effects of extrusion parameters on the fabrication of filament. This study scope will provide more understanding of the influence of elastomer on the PLA in terms of the morphology, thermal properties, and mechanical properties of the TPE filament. Besides, to further enhance the TPE filament, experimental work can consider incorporating a filler.
- Performing 3D printing using the produced TPE filament is crucial to demonstrate TPE filament's integrity under printing conditions to test printability. More innovative research into TPE filament manufacturing with various thermoplastics and elastomers is necessary for 3D printing. Performing experiments with multiple ratios and compatibilizers of TPE blend will create many opportunities for broad manufacturing of TPE filament.
- More research by manipulating FDM variables on the printed part should be conducted, as no research study on TPE material concerning FDM printers are available to date. Besides, more studies focusing on the rheological properties of PLA/elastomer blends should be conducted to get a better understanding of the flow ability of TPE material during the printing process.

PLA-based TPE filament is ideal for flexible goods since it is elastic, flexible, and long-lasting. It also offers a lot of commercial potential for industrial, household, and medical applications, and is also expected to have a strong impact and adhesive characteristics, as well as great dimensional stability. PLA is compostable, and ENR is biodegradable, thus making the usage of TPE filament is eco–friendly and sustainable.

Author statement

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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.

Acknowledgement

The authors wish to thank the Universiti Malaysia Perlis, Malaysia and Lancaster University, United Kingdom for the cooperation and assistance in conducting this research. The authors also would like to acknowledge for the facilities provided by Universiti Malaysia Perlis (UniMAP) and the Ministry of Higher Education (MOHE) of Malaysia for financial support of this research through the Fundamental Research Grant Scheme (FRGS) under a grant number of FRGS/1/2020/TK0/UNIMAP/02/90. The authors also like to extend their great gratitude to Research Management Center (RMC UniMAP), Universiti Malaysia Perlis (UniMAP), for publication incentive grant and all the supports given.

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