Composites: Part A 149 (2021) 106525

Contents lists available at ScienceDirect

Composites Part A

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

Recent developments on the overmolding process for the fabrication of thermoset and thermoplastic composites by the integration of nano/micron-scale reinforcements

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ARTICLE INFO

Keywords:
A. Polymer-matrix composites (PMCs)
A. Moulding compounds
A. Resins
C. Process Modeling

ABSTRACT

Overmolding process is one of the growing advanced technologies for fabricating lightweight composite structures used in the aerospace and automotive industries. This technology enables integrating multiple types of reinforcements from macro- to nano-scale in thermoplastic and thermoset matrices and assembling of dissimilar polymeric materials. Besides, this process is well suited to a digitalization of advanced composite manufacturing for complex geometries with outstanding performance and high adaptation to multifunctionality. The present review aims to cover the recent developments in the design and fabrication of thermoset- and thermoplastic-based composite systems via overmolding process under (i) multi-material injection molding and (ii) insert molding technologies with the employment of nano/micron-scale reinforcements. Multi-material injection molding (or over injection) is investigated by considering two or more thermoplastic polymeric systems in a single mold to obtain high structural performance and bonding quality. On the other hand, the insert molding process is evaluated through matrix and reinforcement types to understand the strength and structural integrity during composite manufacturing. The main bottleneck of adhesion strength in the overmolding process is elaborated with the discussion of distributive approaches and offers a new perspective in producing multi-functional composites in a single-step process with design tools.

1. Introduction

There has been a rising demand for high-performance composite and plastic parts in recent years by applying cost-effective and highly efficient manufacturing techniques. However, the manufacturing cycle times and costs are the main issues for developing multi-functional and multi-scale thermoset and thermoplastic-based composite systems for aerospace and automotive applications. To meet the demands in this field, the overmolding process, a one-shot process, offers to manufacture the composite parts by providing short cycle times in the order of 1–2 min, and adjust time differences between the forming of a polymer part and joining the mold, and control the flow behavior of polymer

Abbreviations: 3DSW, 3D Skeleton winding technology; ABS, Acrylonitrile butadiene styrene; Al_2O_3, Aluminum oxide; CNT, Carbon nanotube; EBC, Ethylene-butene copolymer; EOC, Ethylene-octene copolymer; EPC, Ethylene–propylene copolymers; EPDM, Ethylene propylene diene monomer; FEA, Finite elemental analysis; FRC, Fiber-reinforced composite; FRP, Fiber-reinforced polymer; FRTP, Fiber-reinforced thermoplastic; GNP, Graphene nanoplatelet; G-PET, Glycol modified polyethylene terephthalate; HDPE, High-density polyethylene; IM, Injection molding; iPP, Isotactic polypropylene; LDPE, Low density polyethylene; LFT, Long glass fiber reinforced thermoplastic composite; PA12, Polyamide 12; PA6, Polyamide 6; PA66, Polyamide 66; PAEK, Polyaryletherketones; PBT, Poly(butylene terephthalate); PC, Polycarbonate; PEEK, Poly(ether ether ketone); PEI, Polyethyleneimine; PET, Polyethylene terephthalate; PLA, Polylactic acid; PMMA, Poly (methyl methacrylate); PP, Polypropylene; PPS, Polyphenylene sulfide; PS, Polystyrene; SEBS, Poly(styrene-ethylene/butylene-styrene); SEM, Scanning electron microscopy; SiC, Silicon carbide; SiO_2, Silicon oxide; SRC, self-reinforced composite; srPET, Self-reinforced polyethylene terephthalate; SWCNT, Single-walled carbon nanotube; T_g, Glass transition temperature; T_m, Melting temperature; TP, Thermoplastic; TPE, Thermoplastic elastomer; TPU, Thermoplastic polyurethane; TS, Thermoset; UD, Unidirectional.

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https://doi.org/10.1016/j.compositesa.2021.106525
Received 18 March 2021; Received in revised form 9 June 2021; Accepted 12 June 2021
Available online 15 June 2021
1359-835X/

regarding the part's complexity, and also combine the characteristics of two or more polymeric materials in a single mold without mechanical interlocking or adhesive bonding [1,2]. In general, in the overmolding process, the first material that can be plastic or composite is called a substrate and then covered by injection of subsequent material.

Overmolding can be mainly categorized into two processes: (i) multi-material molding and (ii) insert molding. Multi-material injection involves the injection of the first material, substrate, into the mold, and then the subsequent material is injected onto the solidified substrate [3]. On the other hand, insert overmolding requires a previously prepared insert placed into the mold cavity, and then polymeric material that can be called overmolded material is injected directly on it [4]. Thus, both variants of the overmolding processes promote the cost, process cycle and multifunctionally of the parts by eliminating the assembly without adapting any fastener, flexibility addition to the rigid parts, and lowering the process times via automated manufacturing implementations.

Overmolding technology is widely used in the injection molding process of thermoplastic polymers and employs in the assembly of two dissimilar thermoplastic polymers reinforcing fibers and fabrics. Also, thermoset composite systems can be used as a substrate in insert molding and combine with thermoplastic polymers to attain thermoset-thermoplastic hybrid composite structures. Hence, the achievement of overmolding process with cost-effective manufacturing solutions and short cycle times to manufacture multi-functional parts highly relies on the performance of bonding of dissimilar materials, and prevention of interdiffusion between insert and overmolded materials.

In order to improve adhesive bonding and increase the interfacial interactions between thermoset and thermoplastic structures, the physical–chemical interactions, surface roughness, and wetting degree are important parameters affecting the performance of overmolded parts [5]. In other words, bonding strength and adhesive strength are two influential parameters to attain an ideal overmolded composite by combining thermoplastics, thermosets, and hybrid composites. These hybrid composites' manufacturing via dissimilar materials can be joined by applying three leading joining technologies: mechanical fastening, thermosetting adhesives, and welding. Usage of adhesives is predominantly preferred for bonding application due to lightness and better contact surface. Moreover, adhesive technology has been progressing by employing nano-scale reinforcements. However, each of these additional processes requires additional cost and time.

Lightweighting is one of the essential criteria in automotive and aerospace applications, and thus each weight-reduction strategy to be performed carries significant importance economically and environmentally. The decrease in the amount of primary reinforcement and co-reinforced matrix with nanofillers by developing new compounds for injection molding and new processing technologies like the overmolding process can overcome the needs in the fabrication of lightweight structures. Therefore, overmolding is needed for a comprehensive understanding of design, materials, processing, tooling, and quality control. To the best of our knowledge, there is no comprehensive publication covering all these perspectives to produce an ideal composite structure by using overmolding technology. The present review covers the research progress and classification of the overmolding process by dividing it into two main categories: multi-material injection molding and insert molding. The selection of reinforcement from macro to micron to nano-scale and matrix types (thermosets and thermoplastics) is discussed to understand how uniform assembly gets and produces a high-performance overmolded composite. The review also addresses the design tools to attain ideal mold and flow properties and the moldability of intermediate products. Especially the current review evaluates the performance of overmolding process by discussing different perspectives ranging from interface quality, bonding of dissimilar materials to the compatibility of selected materials with the mold design and flow properties. Therefore, this review summarizes the recent studies in the literature and merges the research tendency in this field with the market requirements.

2. Multi-material injection molding

To manufacture rigid-flexible combinations, the use of the two-component injection molding technology, where the injection molding of a thermoplastic part is combined with the injection molding of an elastomeric component, is becoming more attractive. A high level of automation provides high productivity while guaranteeing a high level of quality [6].

This process is performed by injection of melted polymer over a previous injection, using one or several molds or some extra machining tool as a rotating platen that increases the overall cost. The stepwise processing route is represented schematically in Fig. 1. This process is mainly preferred in three applications: multi-component one-step assembly, multi-color injection molding, and hard-soft combinations [7].

The end product is a single piece fitting together of polymers with distinctive mechanical properties that need to be safe and firmly bonded during its useful life; more than, that it should be separated easily for recycling processes [8]. This technique is preferable to get parts with desired features to raise functionality, including combinations of structural strength and touch sensitivity, vibration and impact tolerance, and cosmetic appearance. Fabrication of durable components overmolded with soft materials is often used to improve the life cycle by preventing shape distortions such as warpage and delamination. While rigid substrate offers a basic framework, soft substrate makes the substance comfortable for consumers [9]. Many different commodity items are used in daily life, such as telephone keypads, toothbrushes, shaving systems, kitchen equipment, hand tools, vehicle interiors, medical devices, and electronic parts in both macro and micron-scale [10].

Table 1 summarizes some of the works related to multi-material and micro-injection by considering the polymer types and reinforcement and their bonding performance. Bonding performance of different material systems in multi-material injection process are evaluated by taking into consideration bond strength related to chemical interactions between polymer chains and interfacial strength indicating physical interactions two bonded/jointed dissimilar material systems at the interface. For instance, Islam *et al.* [11] investigated the effects of injection processing parameters, the addition of glass fiber, surface roughness, environmental factors during the process, and interface temperature on the adhesion performance between different thermoplastic polymeric materials of acrylonitrile butadiene styrene (ABS), polycarbonate (PC), polyethyleneimine (PEI), poly(ether ether ketone) (PEEK) and polystyrene (PS). The results indicated that mold and melt temperatures, melting of polymer surfaces, roughness (for mechanical locking), and annealing above the glass transition temperature (T_g) for the semi-crystalline polymers were found as significant factors to increase the bond strength allowing co-crystallization of two polymers at the interface. Fig. 2 represents the effects of interface roughness on polymer–polymer adhesion and increased bond strength with the increased roughness. Additionally, in thermoplastic processing, the degree of crystallinity is controlled by cooling rates and there is an inverse relationship between crystallinity and cooling rate [20-22]. However, this crucial phenomenon is not particularly addressed for the overmolding process in the literature.

Six *et al.* [12] developed a healing model towards the quantification of the interfacial strength using to define the strength between layers of thermoplastic composites (semi-crystalline high-density polyethylene (HDPE) and thermoset ethylene propylene diene monomer (EPDM)) by the investigation of the parameters of curing and interface temperatures. Experimental results were supported with numerical modeling, and successful plate production was achieved in this study. Furthermore, Arzondo *et al.* [13] attained an improvement in bonding strength between ethylene-octene random copolymer (EOC) and a low-density polyethylene (LDPE) injection molded onto polypropylene (PP) homopolymer with the optimized processing parameters. During the

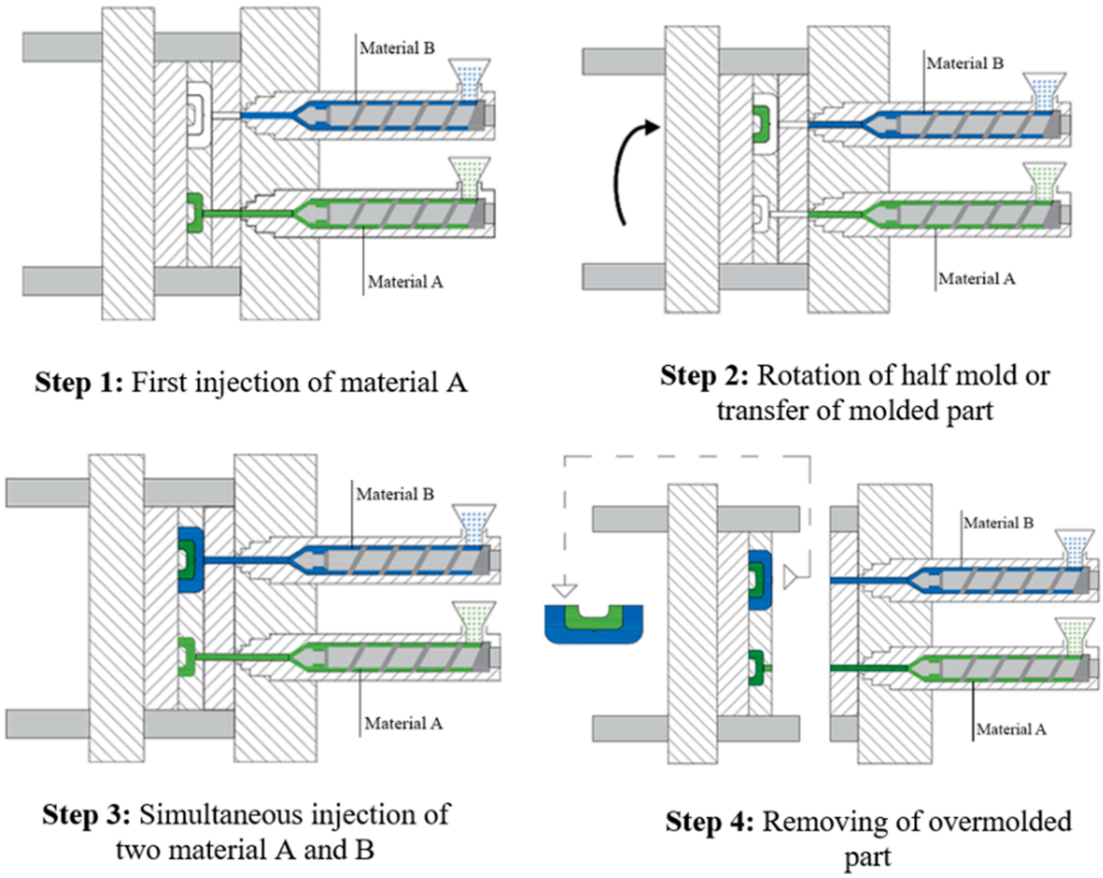

Fig. 1. Schematic representation of the stepwise processing route of multi-material injection molding. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Summary of the studies on multi-material and micro- injection molding techniques.

Method	Polymer Matrix	Insert Material	Improvement	Result	Ref
Multi-MaterialInjection Molding	PC	ABS	Polymer-polymer adhesion and bond strength	An increase in polymer–polymer bond strength by high mold and melt temperatures, surface roughness, and thermal annealing	[11]
	PC	PEI		Improvement in bond strength with higher interface temperature	
	PC	PS		No change in bond strength by addition of glass fiber	
	PEEK	PEI		An increase in polymer–polymer bond strength by thermal annealing	
	HDPE	EPDM	Interfacial Strength between thermoplastic and rubber	Validation of successful numerical strategy by experimental results	[12]
	PP	EOC and LDPE	Bond strength	Attainment of better adhesion with ethylene-octene copolymer (EOC) without melting the PP surface	[13]
	poly(styrene-ethylene/butylene-styrene) (SEBS)Poly (styrene-ethylene/butylene-styrene) (SEBS)	Isotatic PP	Bond strength	An increase in bond strength by higher mold temperature by allowing recrystallization at the surface of insert.	[14]
	TPU	PBT (%30 GF)	Interfacial strength	An increase in bond strength by changing gate design	[15]
	EOC and Ethylene–butene copolymer (EBC)	PP	Interfacial strength	Accomplishment of adhesion bonding with PP inserts by controlling interface temperature An increase in adhesion by 14 mol % octene group	[16]
Micro-Injection Molding	TPE	Aluminum	Optimization of geometry	The effects of mold temperature, melt temperature and injection speed on the geometry of the overmolding material	[17]
	PMMA	Nickel	Filling of cavities at nano-scale	Design and validation of analytical model for filling the nano channels of insert by increasing the mold temperature	[18]
	HDPE	Bulk metallic glass	Filling of cavities at micro/ nano- scale	Improvement in filling of micro/nano features by increasing high holding pressure and high temperature	[19]

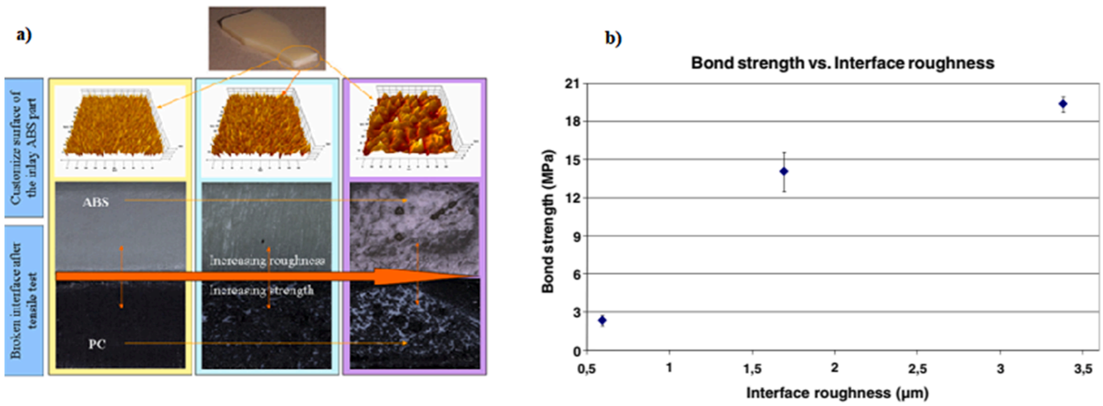

Fig. 2. (a) Effects of interface roughness on polymer–polymer adhesion with increased roughness increased amount of traces of one material visible on the material on the broken surface, (b) Bond strength vs. interface roughness [11]. (Reproduced with the permission of Springer). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

processing, any additional heat treatment was not applied to the interface of a specimen. This led to the minimization of thermal expansion, the prevention of deformation in the parts, and the prohibition of degradation in EOC at high temperatures. Overmolding the EOC on homo-PP is suggested for various applications such as automotive interior parts or household applications. Moreover, Chandran *et al.* [14] enhanced the cohesive tensile strength of isotactic polypropylene (iPP) insert and overmolded thermoplastic elastomer up to 50% at high mold temperature resulting in the migration of plasticizer from an insert to thermoplastic elastomer (TPE) and the recrystallization at the interface. In one of the studies, Frick *et al.* [15] investigated adhesion bond strength by combining 30 wt% glass fiber reinforced polybutylene terephthalate (PBT) as a hard component and thermoplastic polyurethane (TPU) as a soft component and tailored the surface roughness of hard part by applying atmospheric plasma treatment and changing the processing temperature of TPU. In another work, Dondero *et al.* [16] controlled the adhesion between an insert and overmolded materials by using two different random copolymers having different octene content having a direct effect on the interface temperature. Consequently, it is possible to join different polymer systems having different crystallinity degree and melting temperature with multi-material overmolding process and attain an ideal overmolded composite part by controlling physical and chemical interactions at the surface of insert and optimizing process parameters. This will pave a way to fasten scaling up this technology and manufacturing readiness level by providing the combination of different thermoplastic polymers in a uniform structure.

In addition to macro-scale production of multi-material injection molding, micro-injection molding is the type of injection molding process allowing to fabricate complex-shaped parts at the micron scale. This technique can be used to fabricate micro-sized parts of different materials with low tolerances in electronics, biomedical and medical products, automotive industry, telecommunication, and aerospace applications [23]. For instance, Baruffi *et al.* [17] showed the influence of the injection molding parameters such as mold temperature, melt temperature, and injection rate on the geometry of the component produced by overmolding of an aluminum cantilever with the TPE. Controlling the flow behavior of polymer melt directly affects the process and part quality in micron scale. In this aspect, Lin *et al.* [18] modelled the filling behavior of poly(methyl methacrylate) (PMMA) into nano-sized cavities on the nickel-based insert by increasing the mold temperature. Moreover, Zhang *et al.* [19] evaluated the filling of nano/micro-channels and ridges on bulk metallic glass surfaces with HDPE and demonstrated the quality of replications by tailoring the parameters of the flow direction, holding pressure, melting and mold temperatures. Consequently, both macro- and micro-scale multi material injection molding allows the creation of multi-layered structures by bringing the polymers having different characteristics into a single plastic structure and it is also possible to get high degree of bond strength and interfacial adhesion with the optimum process parameters and adjusted surface roughness of the prepared inserts.

3. Insert overmolding

There are several attempts to develop hybrid composites by the overmolding process to produce lightweight structural composite parts, especially for thermoplastic polymer-based composites [24]. The insert overmolding process includes the placement of the fiber-reinforced composites or pre-molded structures as an insert in the mold prior to the injection of the second material [25]. In some variants of this process, the insert is preheated below the melting temperature, and then the second polymeric material (overmolded material) is injected subsequently on this insert [15,26]. The insert molding can also be achieved by injecting the polymer directly onto the substrate without applying any preheat treatment. Then, during the cooling process, a packing pressure is applied after the injection process to eliminate the thermal shrinkage of the resin. Fig. 3 shows the schematic representation of the insert overmolding process steps. After the shaping of inserts (Step 1), the pre-shaped insert is placed into the mold cavity, the surface of the insert is heated, and mold is closed (Step 2). Process continues by overmolding (Step 3) and completes by removing of finished part (Step 4). In step 2, the interface is usually heated up thermally by the heat of the injected liquid polymer to bond the insert and injected polymer. The available time for bonding of melt polymer and substrate is limited due to the low mold temperature for rapid cooling, allowing short cycle times (<1 min cycle time) [5,25]. Successful joining/bonding of an insert and an injected polymer within this limited time frame can be provided to investigate two complementary phenomena; i) development of intimate contact between an insert and an injected polymer and ii) interdiffusion of polymer chains across the interface [5]. Bond strength is important to prevent the separation of layers and thus resin selection carries a significant importance to control the bonding mechanism involving in chemical and physical interactions. In overmolding process, mostly thermoplastic resin systems are preferred over thermosetting resins since thermoset based composite production requires a long curing process, and it can be time consuming. On the other hand, it is possible to combine the thermoset and thermoplastic materials within one part in overmolding process resulting in strong interface. Carbon fiber-based epoxy composites can be used as an insert and the appropriate surface pretreatments such as atmospheric plasma, oxygenation and chemical activation applied on a thermoset based insert provide to form sufficient interface strength with the injected thermoplastic resin.

In literature, the insert molding is generally investigated in terms of

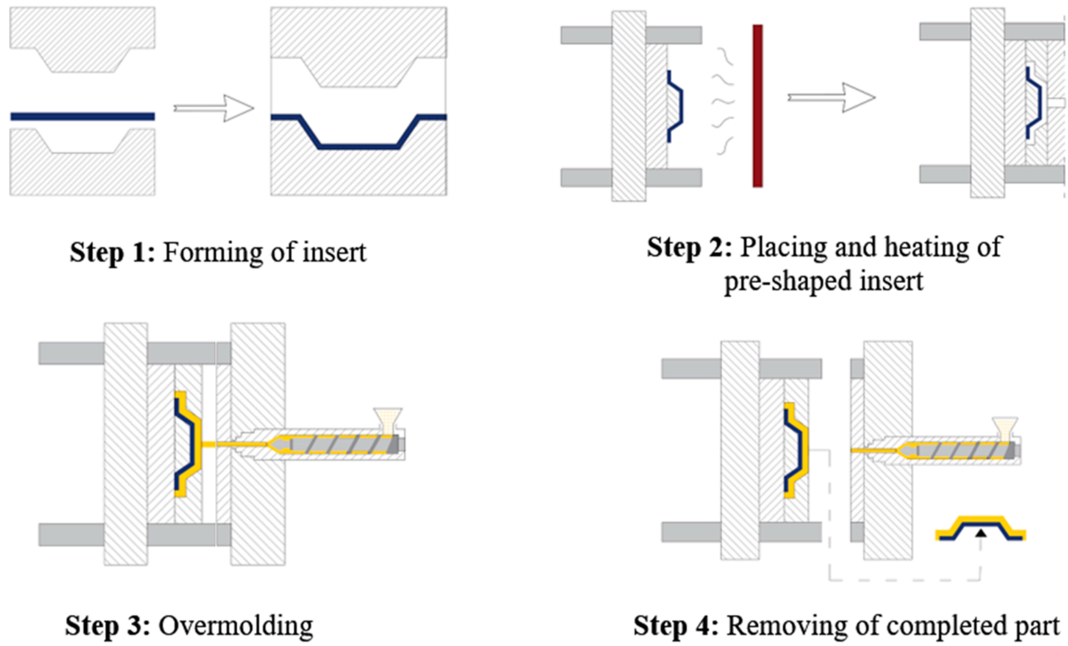

Fig. 3. Schematic representation of step-by-step insert overmolding process (Shaping of insert (Step 1), placement of insert into injection mold and heating the surface of insert (Step 2), overmolding onto insert (Step 3), and removing the solid part (Step 4)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the matrix and fiber types and the process parameters resulting in improvements in different behaviors, summarized in Table 2. According to these published studies, the main issue in the overmolding process is the inefficient adhesion strength between the insert and overmolded materials. Therefore, the relation between the selected matrix and reinforcement carries significant importance to produce an ideal structure, and the adjustment of process parameters is essential to increase the compatibility of the chosen material systems. There are various reinforcements used in overmolding processes, such as organosheets, UD tapes, fabrics, and laminated structures. The adaptable process is needed to be optimized to enhance the interfacial interactions regarding the type of matrix that can be thermoplastic, such as engineering and high-performance plastics or thermoset polymers like epoxy.

3.1. Types of polymeric matrix used in insert overmolding

The type of polymer matrix has a significant influence on the process parameters and the target application. There is an increasing trend in replacing thermoset composites with thermoplastic composites due to environmental issues, recyclability, and higher moisture absorption rates [37]. Recent studies for different applications focus on the improvement of thermoplastics (TPs) applications and the development of their composites. Specifically, organosheets which are high performance and semi-finished materials consisting of carbon, glass, or aramid fiber fabrics embedded in thermoplastic resin, have been drawing attention in manufacturing thermoplastic structural components due to their advantages of shorter process times, better impact strength, easier formability, the formation of void-free structure, and improved recyclability in comparison to conventional fiber reinforced thermoset (e.g. epoxy) composites. In the insert overmolding process, pre-impregnated fiber reinforced sheets are first heated and shaped in the injection mold to form a thermoplastic composite, and subsequently, the thermoplastic material is overmolded onto the composite. Generally, the matrix of organosheets and overmolded thermoplastic are compatible with each other providing an acceptable adhesion performance [38]. However, thermoplastic composites have some drawbacks, such as the high cost of high-performance polymers (e.g., PEEK, PAEK polymers) and limitations in thermal stability especially engineering plastics (e.g., PA, PP, PET) [39,40].

Structural composites with a thermoset (TS) matrix perform well in applications where thermal resistance and strength are required [41]. Compared to thermoplastic, the crosslinking of the thermoset polymers restricts the reshaping [42-44]. As mentioned before, preheating of the insert is preferred in some insert overmolding techniques since it affects the diffusion of polymer. Despite some advantages of using TS, creating designs with built-in assembly and connection details has been problematic. Besides that, the lack of welding options and difficulties in complex shapes, particularly with the technical composites, are worth noting as other disadvantages of using thermosets [30].

3.2. Types of reinforcements used in the overmolding process

Fiber-reinforced thermoplastics in a laminate form (organosheet) are used as an insert material of hybrid composite parts. Various textile structures, including unidirectional (UD) fabrics and woven or nonwoven fabrics, are commonly available for the organosheet [45]. Preheated organosheets are inserted into the mold of IM and thermoformed to a specific shape. Moreover, thermoplastics reinforced with short or long fibers (e.g. discontinuous fiber) are injected onto the inserted parts by IM for structuring multiple parts [1,46,47]. Due to the integration of optimal processes for each material, this methodology provides new ultra-lightweight parts with high mechanical properties, recyclability, and short-cycle manufacturing [33].

The motivation towards adapting the composite materials is to make lighter parts without sacrificing their mechanical strength and structural integrity [48]. Despite the categorization of composites based on matrix type, they can also be divided according to the type of reinforcement [49]. Fiber-reinforced composites (FRCs) are mostly used to address the advanced composites, constituting the greatest amount [50]. The fiber reinforcement of thermoset or thermoplastic matrix can take place in different forms (such as long, short, yarn, mat), and the fiber materials are mainly carbon fiber (CF), glass fiber (GF), natural fiber (NF), or thermoplastic fibers [51,52]. The dimensions and types of fiber reinforcement playing a crucial role in the performance of the overmolded process are further discussed in the following section according to the type of fiber type.

Table 2
Summary of the recent studies on insert overmolding process.

Matrix		Insert			Investigated Parameters	Improvement	Result	Ref
Resins	**Fiber**	**Form**	**Fiber Type**	**Resin**				
PP	Glass fiber	Laminate	Glass fiber (woven)	PP	Preheating temperature	Welding strength	– Improvement in the welding strength with increasing of melting temperature and holding pressure – A decline in the welding strength with decreasing mold temperature due to higher residual stress	[26]
EPC and EPDM	x	Plate	x	Homo PP	Chemical compositions and microstructures of EPC-PP melt temperature	Adhesion strength	Better penetration between ethylene propylene copolymer (EPC) and PP	[8]
PC/PET	x	Self reinforced-sheet	PET fiber	PET	Temperature	Tensile strength	Reduction in tensile modulus due to relaxation of residual stress above T_g of the matrix of the insert	[27]
PP	x	Plate	Short Glass Fiber	PA 12	Effect of powder sintering (PA12)	Adhesion	Improvement in adhesion by applying powder sintering	[28]
PEI	Short GF	Sheet	Carbon Fiber	PEI	Part geometries	Interfacial strength at high temperature	Optimization of part geometries for the interface characterization	[29]
PA 6	x	Fabric	Carbon Fiber and Glass fiber	Epoxy	Fiber Type, molding temp and roughness	Adhesion at interface	Better adhesion for the roughened carbon fiber processed at high temperature	[30]
PP	Long glass fiber	UD tape	Glass fiber	PP	Process parameters	Experimental and FEA of bond behavior	Improvement in the bond strength due to enhanced diffusion at the interface	[31]
PLA	x	Mat	Jute fiber	PLA	Fiber orientation and content of fiber/polymer	Increasing flexural strength and thermomechanical resistance	Improvement in the mechanical properties due to high content of fiber oriented in $-45^\circ/+45^\circ$	[32]
PP	Chopped Glass fiber	Laminate	Short carbon fiber	PP	Effect of CNT on the interfacial strength	Increasing interlaminar shear strength	Increase in the shear strength by addition of CNT (1 wt%)	[33]
PET and G-PET	x	Self reinforced-sheet	PET fiber	PET	Types of matrix	Tensile and impact strength by enhancing adhesion	Attainment of higher mechanical properties by using G-PET in comparison with PET	[34]
PA6	x	Plate	x	PA6, AP-Nylon ® based and Mg catalyst in-situ polymerization	Melting and mold temp., holding pressure and time	Bond strength of preform	– Improvement in the bond strength with increasing melting temperature – Weakening in bond strength with increasing holding pressure and time	[2]
PA6	x	UD Tape	Carbon fiber	PA6	Temperature	Shortening process route by preheating	– A decrease in processing time by preheating – Improvement in mechanical properties	[15]
PP and PPS	Glass Fiber	Yarn	Glass fiber	PP	The fiber content in matrices	Increasing elongation at break	An increase in breaking forces by using continuous fibers	[35]
PP PEEK	Glass fiber Carbon fiber	Laminate	Glass fiber Carbon fiber	PP PAEK	Mold temperature, injection temperature and insert temperature	Strengthen interface bonding	Optimization of process parameter by simulation and mechanical tests with high temperature	[25]
PP	Short glass fiber	Laminate	PP	Continuous carbon fiber	Working temperature	Interfacial behavior	A decrease in interfacial shear strength and shear stiffness with increasing working temperature	[36]

3.2.1. Glass fiber-based overmolded composite production

Glass fiber (GF) is a kind of natural ore made of soda ash and boric acid. The molten state is drawn or blown into a very fine fibrous material by external force [53]. GF has high specific strength, excellent high-temperature resistance, corrosion resistance, chemical stability, electrical insulation, and thermal insulation properties [53,54]. GF combined with different matrices can exhibit different characteristics, properties and can serve different applications. There are several studies on glass fiber-based overmolded composite processing and their performances. For instance, Giusti and Lucchetta [26] overmolded a laminate form substrate to prepare a T-joint specimen by adjusting the process parameters. PP laminate was reinforced with woven glass fiber fabric (50 wt%), and overmolding materials were reinforced with short glass fiber (30 wt%). The results demonstrated that melt temperature and holding pressure-assisted increase the welding strength by promoting an intimate contact between two layers and diffusion of polymer chains. Mold temperature was considered as another significant parameter for materials having high melting temperatures. With decreasing mold temperature, welding strength was decreased because of high residual stress at the interface, which resulted from the high amount of shrinkage. Moreover, selective preheating of the substrate was suggested instead of heating of whole part. In another study, Ott *et al.* [28] used polyamide 12 (PA12) as local additive structuring to generate media-tightness between insert and PP as an overmolding material. Using an infrared (IR) spot, the surface of the insert composed of PA12/short GF was melted, and a powder layer made of PA12 was applied. Improvement in both bond quality and media-tightness by the additive structuring was observed clearly, as presented in Fig. 4.

Moreover, Joo *et al.* [31] investigated the bond behavior of the composites, which are previously studied by Wakeman *et al.* [55], according to overmolding process parameters. The bond behavior of a continuous glass fiber-reinforced thermoplastic composite rod (3D-Tow) overmolded with long glass fiber reinforced thermoplastic composite (LFT) given in Fig. 5 is investigated. The structural simulation was made to predict the mechanical behavior of the prototype front bumper, and tensile tests are carried out experimentally. The results revealed that interfacial strength was enhanced at a high injection flow rate and molding pressure due to the diffusion at the interface. Furthermore, Akkerman *et al.* [25] inserted a rod, fabricated by laminating continuous glass-fiber reinforced thermoplastic tape, into the mold cavity then it is overmolded with the long glass fiber-reinforced PP to decrease the high cost of some processing methods such as hot compression molding. The work conducted by Beck *et al.* [56] presented a 3D skeleton winding technology (3DSW) manufacturing process and developed the 3D test specimen obtained by connection of three main components. Fundamental investigations of these structural components with overmolded fiber skeletons demonstrate the potential of continuous fibers in injection molded components made from PP and polyphenylene sulfide (PPS). Therefore, different forms, as well as sizing chemistry of glass fibers, directly affect the mechanical performance and compatibility with the selected matrix for the fabrication of overmolded composites in different shapes.

3.2.2. Carbon fiber-based overmolded composite production

Since the 1960s, carbon fibers have become one of the most important industrial materials. Carbon fibers are high-strength and high-modulus fibers with a carbon content of more than 90%. At present, the modulus of carbon fibers can reach 90–95% of the perfect graphite modulus (1025 GPa) [56,57]. It is resistant to high temperatures, friction, electrical conductivity, heat conduction, and corrosion [58-61]. Also, carbon fibers with high formability can be processed into various fabric forms. However, the carbon fiber reinforced composites with excellent properties also require excellence in adhesion at the fiber/matrix interface, determined by the chemical and physical interactions at the interface [62,63].

There are different attempts to use carbon fibers or both carbon and

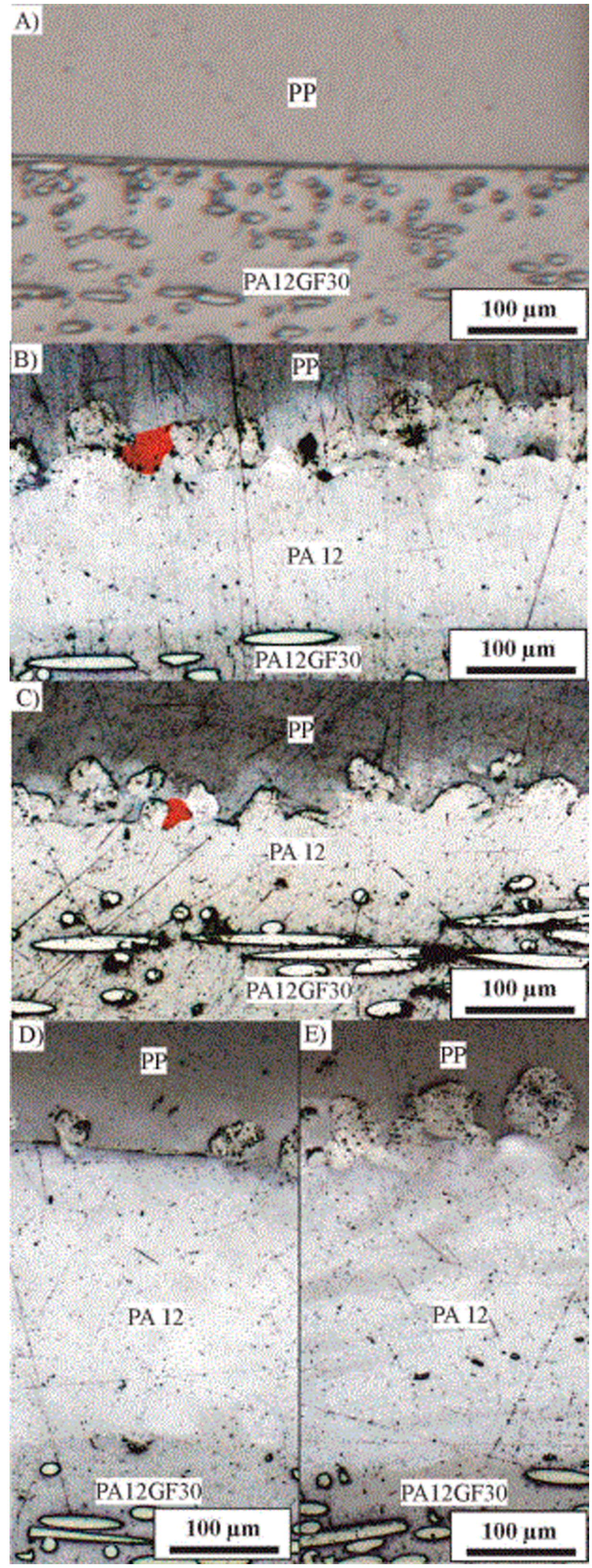

Fig. 4. Microscopic images of reference part (a), one layer without force (b), one layer with force (c), two-layer without force (d), and (e) two-layer with force [28]. (Reproduced with the permission of Elsevier). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

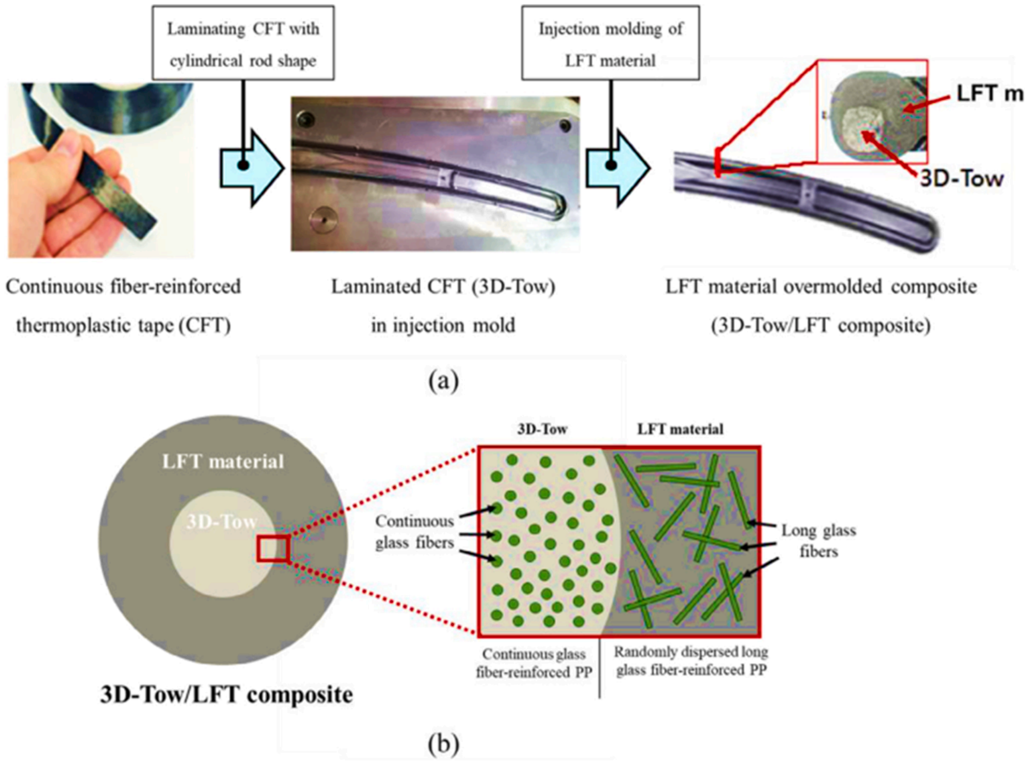

Fig. 5. (a) An automobile component made by 3D-Tow/LFT composite, and (b) schematic representation of 3D-Tow/LFT composite [31]. (Reproduced with the permission of Elsevier). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

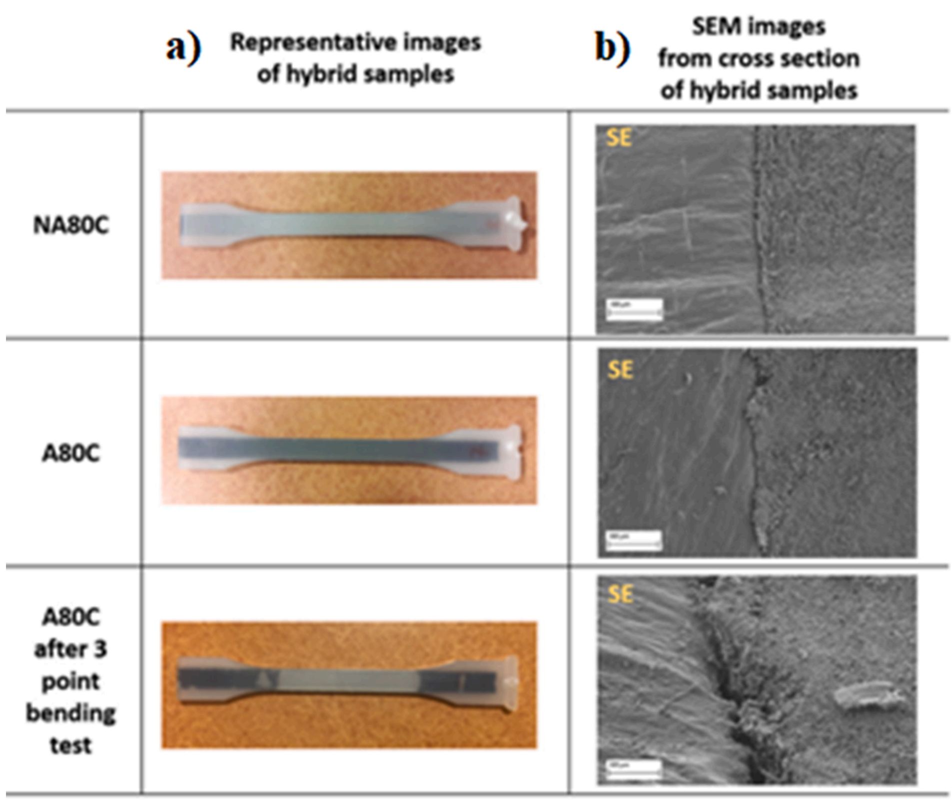

Fig. 6. Photographs and SEM images of cross-sections of specimens (a) before and (b) after 3-point bending test [30]. (Reproduced with the permission of Elsevier). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

glass fibers in manufacturing overmolded composite structures in industrial applications and academic studies. Karakaya *et al.* [30] investigated fiber types' parameters, mold temperature, and roughness affecting adhesion between over-molded polyamide 6 (PA6) and continuous glass and carbon fiber-epoxy reinforced composites. The presented results indicated that the peel ply treatment and heat treatment at high temperatures (80 °C) significantly improved the interface's bonding strength. The carbon fiber side with higher roughness showed better adhesion compared to the side of glass fiber due to the differences in surface compositions. The photographs and SEM images of specimens before and after the 3-point bending test are represented in Fig. 6. The color difference between non-roughened (NA80C) and roughened (A80C) specimens points out compatibility between layers. The dark color is the demonstration of improved adhesion which is supported with SEM images.

Moreover, Joppich *et al.* [29] designedtwo different molds by optimizing the part geometries to investigate the effect of the interfacial strength on the performance of overmolded composites. They monitored the welding conditions between the samples of polyethyleneimine (PEI)/CF organosheets overmolded and PEI reinforced with short GF. Additionally, a simulation model was developed to predict welding temperatures and pressures for the improved adhesion in the hybridization of PEI-based materials by the overmolding process. Fu *et al.* [36] worked on constitutive modeling of a hybrid composite of carbon fiber reinforced laminate overmolded with glass fiber reinforced PP to observe temperature-dependent interfacial behavior. Decreasing both interfacial shear strength and stiffness of hybrid composites with increasing service temperature ranged from 23 to 90 °C was found. Consequently, the utilization of carbon fibers in the hybrid composites fabricated by insert overmolding has been advanced by accomplished adhesion due to the significant impacts of carbon fibers on performance.

3.2.3. Natural fiber-based overmolded composite production

There is a growing tendency in the natural fiber (NF)-reinforced composite market to fabricate bio-based composites and structures. NFS as reinforcement agents are the environmentally friendly candidate among the other fiber types and can be naturally degraded in the environment. Today, there is a growing tendency to use natural fibers such as flax, hemp, jute, kenaf, and sisal in bio-based composite manufacturing [64]. These fibers can reduce the cost and provide an improvement in the toughness, specific strength, and modulus of the selected polymer matrix at high loading percentages [65,66]. On the other hand, NFs show lower strength (140–2000 MPa) than synthetic fibers as glass or carbon fibers (2000–4000 MPa) and also have some drawbacks such as high moisture absorption rate, poor fire resistance, variable quality based on harvesting condition and limited processing temperature [67-69]. Especially moisture absorption behavior of NF polymer composites can cause a decrease in the mechanical performance resulting in microcracks at fiber–matrix space, and also a change in dimensional properties of composite structures and this stems from the hydrophilic nature of cellulosic structure having high hydroxyl groups available in all NF fibers [70,71].

Although the aforementioned drawbacks of NF do not meet the standards in structural parts, especially in the aerospace, defense, and automotive industry, NFs can be found more potentially in everyday products subjected to moderate loadings. With the growing environmental crisis, there is a tendency to fabricate fully green composites of natural fibers with biodegradable resins for industrial composite production [72]. Therefore, there are several works in the literature to demonstrate the potential performance of NFs in the composite structure. In one of the studies, Wis *et al.* [32] fabricated environmentally friendly, biodegradable composites (named ecosheets) by overmolding of poly(lactic acid) (PLA) onto PLA/jute-mat, reinforced continuous fiber composite sheets, and investigated the effect of the fiber orientation and mechanical properties. Fig. 7 shows the schematic representation of the production process of ecosheets by overmolding and SEM investigation of fractured surfaces, indicating that there is still an adhesion problem at the interphase. However, mechanical properties were increased (at the maximum value, the strength of ecosheets was obtained as 130% compared to neat PLA). In another work, Andrzejewski *et al.* [73] produced a laminate consisting of PLA/flax by using compressing molding and then performed injection molding with neat PLA and PLA having 20 wt% harl particles coming from flax fiber waste resulting in no change in heat resistance properties but an improvement in the stiffness and impact strength of molded parts. This study shows that NF based PLA composites having the relatively low glass transition temperature initiate the surface interdiffusion without any additional pre-heat treatment step of insert before injection and thus leading to the reduction in the energy consumption and cycle time.

To conclude, the growing rate of natural fiber consumption in composites is not in the expected range, and the market is slowly growing due to the limitations in mechanical performance and surface properties in natural fibers-based tapes and prepregs. In order to enhance the compatibility of NF with the selected matrix, there are several studies using physical or chemical surface modification methods such as plasma treatment, mercerization, silanization acetylation, and permanganate treatment [74-76]. The selection of suitable surface modification of NFs can strengthen fiber/matrix interactions and improve interfacial adhesion between an insert and overmolded parts. On the other hand, each additional treatment for these bio-based fibers

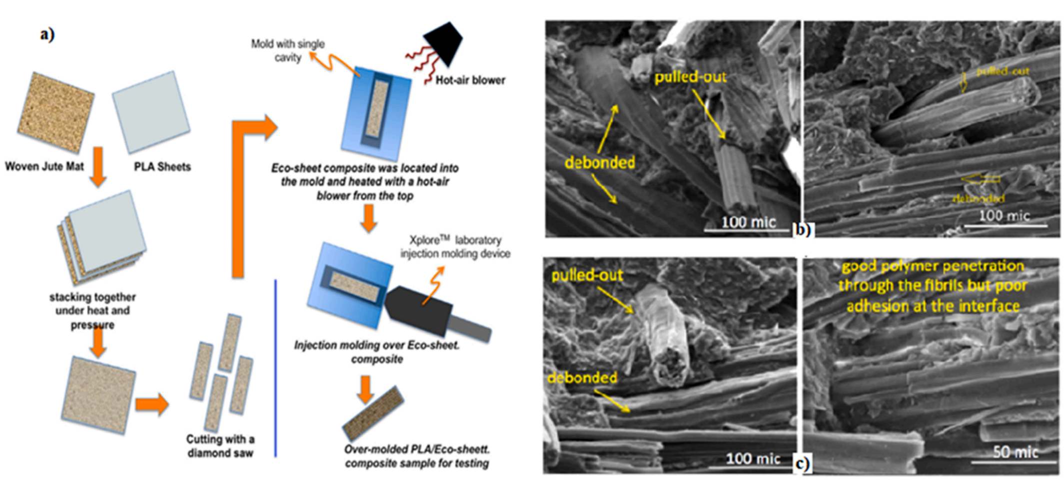

Fig. 7. (a) Preparation route of ecosheet by insert overmolding and SEM micrographs of fractured surfaces after (b) flexural and (c) tensile tests [32]. (Reproduced with the permission of Wiley). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

might lead to an increase in cost.

3.2.4. Self-reinforced overmolded composite production

Self-reinforced composites (SRCs), or single-polymer composites, in which a polymer matrix is reinforced with fibers, tapes, and/or particles of the same polymer, are one of the promising implementations of the insert overmolding process. Maximum compatibility between matrix and reinforcement agent with higher relative homogeneity can be achieved in the SRCs [66,72]. The characteristic property of this type of composites is their full recycling possibility since the same polymer is utilized for both matrix and reinforcement comparison to the composites consisting of different classes of components. In other words, self-reinforced composites can be re-processed by melt techniques without applying any additional separation techniques [34]. Despite their advantages, SRCs have a fundamental limitation regarding the cost of production and complexity of the processing cycle. Compression molding is a widely used technique for the processing of SRCs, falls behind the highly efficient and flexible manufacturing techniques such as injection molding [77,78]. Usage of self-reinforced materials for injection molding technologies especially insert overmolding is very limited. In one of the studies, Andrzejewski *et al.* [34] fabricated self-reinforced inserts made from self-reinforced poly(ethylene terephthalate) sheets (srPET) overmolded by the matrix polymer (PET and G-PET) with the standard injection molding process parameters. The results confirmed that the composites made from glycol-modified PET (G-PET) were favorable with an increase of 60% and 32% in tensile strength and elastic modulus, respectively. Fig. 8 shows the high level of adhesion between matrix (PET and G-PET) and self-reinforced sheet at the fracture surface examined by SEM, and the tearing of fibers oriented in parallel can be observed in the corresponding SEM images of specimens.

Overmolding of srPET was also studied by Jerpdal *et al.* [27] that reduced both weights of the automotive component and environmental impact compared to glass-reinforced thermoset composites. The effects of temperature on mechanical properties of composite fabricated by overmolding with polycarbonate (PC)/PET onto srPET during the processing cycle were investigated comprehensively. Mechanical strength was decreased by 18% due to the residual stress that occurred during the processing of composites. To conclude, self-reinforced composites should be considered for recycling and better adhesion in the new design of composites and are needed to improve their mechanical performances by tailoring process parameters.

3.2.5. Nano-scale reinforcements in bonding systems

Hybrid composite structures combining thermoset-thermoset, thermoplastic-thermoset, thermoplastic-thermoplastic, and thermoplastic-metal parts have taken great attention due to growing demand for the replacement of all-steel structural parts, especially in automotive and non-automotive applications such as household appliances and bicycles [80]. At this point, interfacial roughness and interface properties become a critical parameter in the overmolding process to increase the compatibility between two dissimilar materials [81]. Herein, adhesive technology represents an alternative to traditional mechanical joining methods such as welding and bolting. Adhesives become prominent due to their lower weight, lower fabrication cost, greater contact surface area between the adherents, and the consequently uniform stress distribution in the bonded region by adding a small amount [82]. In order to attain the required level of adhesion strength between the overmolded material and the surface of the insert, the adhesive formulation is needed to develop by integrating different kinds of nanomaterials since adhesives are suffering from low mechanical and thermal properties resistance and need additional surface preparation steps.

With the development of material science, hybrid joints obtained by nanofillers' addition are getting attention owing to their significant enhancing features such as mechanical properties, bonding strength, electrical conductivity, and thermal properties [83,84]. Adhesives reinforced with nano-scale particles might serve as an intermediate shock-absorbing elastic layer between the two stiff components and increase the bond strength [85]. Moreover, the reason for significant improvement in mechanical properties can be explained by the occurrence of local shear bands, which generate stress concentrations around nanoparticles during applying loads [86]. The optimum filler level to maximize the bond strength might be affected by several factors, including the size, shape, and content of the filler particles, the surface properties of the filler. Among these, the uniform dispersion of nanofiller in the adhesives is a notable factor improving adhesion behavior. The functionalization of the nanoparticle surface can be utilized as an alternative solution to enhance chemical interactions between nanofillers and adhesive.

There are various nano-scale reinforcements such as carbon nanotubes (CNTs), graphene nanoplatelets (GNP) [87-89], nano-clay [90], nano-silicon oxide (SiO_2), nano-silicon carbide (SiC) [91], nano-aluminum oxide (Al_2O_3) [92] and others [59,85,93] widely incorporated into adhesives to improve the interfacial interactions during co-bonding of similar and dissimilar materials by controlling thermodynamic affinity and physical interactions [94]. For instance, Li *et al.* [86] achieved increased shear strength up to 133.2% by the addition of 1.0 wt % functionalized SiO_2 with dendrimer containing reactive amino groups in the adhesive system. Herein, surface functionalization was carried out to solve the poor bonding problem between SiO_2 and epoxy. In another study, Zamani *et al.* [95] incorporated silica and GNP into commercial epoxy-based adhesives to investigate the fatigue life of composites. The results revealed that combination nanofillers (0.5 wt% of each particle) in the adhesive improved the fatigue life. The addition of more than 1.0 wt% of nanoparticles can cause an agglomeration problem and thus reduce mechanical properties. CNTs are preferable nanofiller due to their excellent tensile strength and electrical conductivity [96]. A combination of mechanical and electrical properties of individual nanotubes promotes them for their utilization as reinforcing agents in

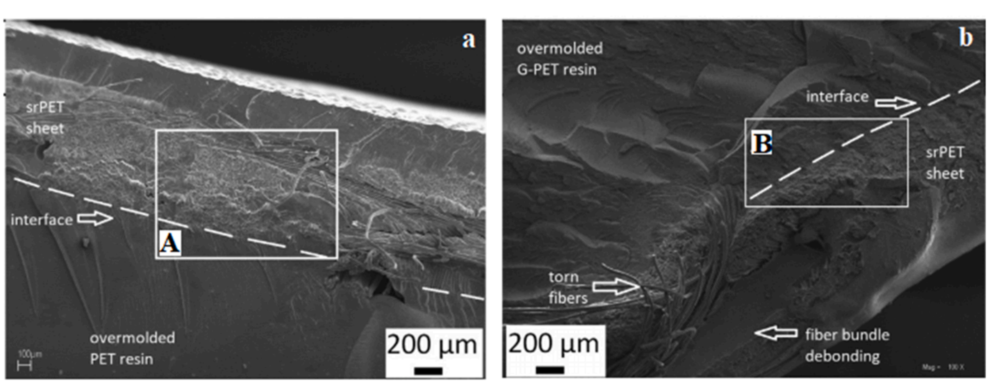

Fig. 8. SEM images of fracture surfaces of self-reinforced PET sheet overmolded with (a) PET and (b) G-PET [34]. (Reproduced with the permission of Elsevier).

several adhesive applications for the aerospace industry, especially. In one of these studies, Jakubinek *et al.* [97] filled epoxy adhesive with a single-walled carbon nanotube (SWCNT) (0.5 and 1 wt%) to investigate the effect of CNTs on the electrical conductivity and joint performance. While the conductivity and peel strength were improved, lap shear strength was reduced in components prepared with 1 wt% SWCNT.

Although various studies revealed nanofillers' influence on the enhancement of bonding strength, the integration of these nanofillers into the overmolding process has not yet gotten much attention due to the obstacles in their high cost, distribution, surface composition, and dimension. Therefore, the studies combining the adhesion technology and the overmolding process with the nanoparticles are limited, especially in scalable manufacturing technologies. There is one particular research where CNTs (0–3 wt%) were utilized in the film form with the carrier of PP polymer to enhance the interfacial strength between UD carbon fiber reinforced laminate and short carbon fiber reinforced PP in the overmolding process [33]. Therefore, nanomaterials with different aspects and functionality have great potential to be adapted at the insert materials by their incorporation into adhesive film with thermoset and thermoplastic systems or other forms and surface treatment tools such as spraying, plasma treatment, and chemical or physical surface activation.

4. Design tools for the overmolding process

The creation of design tools to fabricate components represents one of the most time- and cost-consuming phases in the development of new products. Design tools have to be developed and integrated into the manufacturing process to reduce the manufacturing lead time and cost [98]. New design tools help improve part manufacturability, minimize manufacturing defects, and reduce overall production time by improving process efficiency. However, the creation of design tools to fabricate components represents one of the most time- and cost-consuming phases in the development of new products. Although the feasibility of the overmolding process has been increased frequently in recent years, there is a lack of comprehensive and complementary design tool strategies.

There are three complementary stages for the development of a design tool framework for the overmolding process: i) process selection, ii) part and process design, iii) mold design. The selection of an appropriate overmolding process (multi-material or insert overmolding) is the initial step to get an ideal designed product by considering labor costs, available equipment, and material types. While multi-material injection could be operated for a higher volume of production and moldable substrates, insert overmolding is preferable for complex substrates such as composite materials or metals. In the insert overmolding process, some additional processes such as shaping and heating the surface of the substrate should be required to get the desired shape. For instance, Moll *et al.* [35] designed and modeled a gripping system allowing further heating and performing UD laminates composed of PA6 reinforced 60% of carbon fiber. In this work, the calculation of temperature loss during transportation was used in modeling to optimize the heating temperature of the insert surface. Achieving maximum adhesion between the substrate and overmolding materials is the main challenge for each overmolding technique. Compatibility between polymeric materials is the main parameter to attain better chemical bonding. However, material selection can be complicated due to complex processing in comparison to injection molding. Thus, the selection of the overmolding process necessitates the consideration of the multidisciplinary physics in a compressive manner with the bonding strength as the governing phenomena.

Similarly, the successful bonding performance of overmolded material is also a leading criterion for the second stage, part, and process design. In part design, uniform wall thickness is critical for ensuring the bonding. The wall thickness of overmolded material should equal to or less than the wall thickness of the substrate to prevent the warpage problem from shrinkage caused by residual stress [35]. Mechanical interlocking, which is another way to bond layers of substrates and overmolding materials, is highly affected by part geometry and surface roughness. Moreover, the warpage problem arising from the heating and cooling profiles during the process is under the consideration of the part design. After the second injection (overmolding of the substrate), a shrinkage problem can occur in the finished product since shrinkage is highly dependent on the type of material, part, and mold design, as well as process parameters such as cooling and heating [99]. The shrinkage issue can be eliminated by adjusting the process and material parameters during the overmolding process [100,101].

In process design, process model and optimization are essential for both cost and time-saving processing compared to trial and error-based manufacturing for the process parameters. The process parameters are critical as the material selection, the type of substrate, and the reinforcements materials to obtain a high-performance composite. Injection speed, melt temperature, mold temperature, packing time, packing pressure, cooling time are the typical injection molding process parameters affecting the overmolding process. Among these, injection pressure, melt and mold temperature, cooling time, and substrate temperature are considerable due to their influences on the interface temperature. Molten state of overmolded polymer paves the way for the intimation of the surface. In addition, the time is required to achieve full intimate contact and fast cooling time to shorten the cycle time. To achieve optimal bond strength, higher than normal melt temperatures are often required. Additionally, the heating of substrate favors the bond strength due to the fact that a thin molten layer at the surface would increase the interaction of polymer chains and posse to co-crystallization during the interface forming. However, these kinds of additional preheating process will result in to rise in cycle time and cost. Thus, a process design should encounter the tradeoff between accuracy and efficient computational effort.

After the optimization, the accuracy of the simulation tool should be validated with experiments and analysis. Additionally, the output of the simulation tools highly depends on the accuracy of the material properties. Thus, a robust simulation tool requires well-established numerical approaches and accurate material properties affecting the process. Prediction of distortions by simulation software can be used to avoid the deformation, matrix cracking, or delamination problems that result from the non-uniform distribution of residual stress arise from weak bonding between surfaces [102]. For instance, Fetecau *et al.* [103] designed an accurate fabrication of part by Autodesk Moldflow software to investigate the flow behavior and effect of interface geometry on the bonding combination of three different materials (e.g., LDPE, HDPE, and PP).

Mold design is the last stage of the designing tools of the overmolding process. One of the main problems of the insert overmolding technique is the placement of the insert in the mold, and the insert should be fixed and stabilized against the injection pressure. Furthermore, flashing problems can be observed at the end of the molding process due to the displacement of the insert. Gate design and location, and configuration of runners, which can also be integrated with the part and process design stage, have also affected the quality of the end product. Despite these aforementioned requirements, researchers have mainly been considered enhancing the interfacial strength by controlling the injection process parameters as melt temperature, pressure, mold temperature, substrate temperature, and cooling time [2,11,31,104]. Key issues are the predictability of the process-induced shape distortions of the overmolded part and the bond strength between the overmolded insert or substrate and injected polymer resin. However, both part/mold design and process optimization largely depend on experience and a trial-and-error process because of the absence of an available design tool to predict interfacial strength.

5. Potential application and outlook

The present review provides a comprehensive study about the recent developments in the overmolding process to fabricate polymer-based composite by incorporating nano/micron-scale reinforcements. This overmolding process comes up with the advantages of cost and time effectiveness by eliminating additional assembly processes. Multi-material injection molding and insert overmolding techniques have been utilized as types of overmolding processes. The multi-material injection has mostly been preferred for esthetic and ergonomic requirements. Due to the availability of numerous polymer materials for multi-material injection molding, the performance of bonding between different polymers has been evaluated through empirical observation, especially in industrial applications. On the other hand, the insert overmolding process shortens process cycle time without extra machining cost to produce lightweight and strengthen composite. Moreover, overmolding can be adaptable from a medium-scale to a large-scale production process.

Despite the differences, both two techniques have been suffering from weak adhesion at the interface. Although some factors such as mold temperatures, annealing, and melting temperatures are adjusted to enhance the bonding of different polymer systems at the interfaces, shape distortions and delamination are observed in the produced overmolded parts. Herein, nanomaterials carry a significant potential to increase surface roughness and physical–chemical interactions allowing better bonding between an insert and overmolded materials. However, nano-integration is at the developing stage in the overmolding process, and there are few attempts to combine nano reinforcements with commercially available continuous fibers such as glass, carbon, and natural fibers. If the synergistic effect is achieved by bringing different scale reinforcers in a uniform structure, an ideal overmolded part will be produced by eliminating current process and manufacturing problems. In order to enhance the interfacial interactions in overmolded parts, the degree of roughness on the surfaces should be increased to provide interlocking of the substrate and overmolding material. On the other hand, there are some attempts to get an ideal wettability degree for the chemical interactions by optimization of injection molding process parameters such as mold temperature, mold pressure, and melting temperature.

In recent years, there is a tremendous interest in the replacement of thermoset and metal composites with thermoplastic materials in terms of lightweightening, sustainability, and circular economy issues. Overmolding technology can cover the prime focus of the industry's needs on waste and energy consumption reduction, recycling, alternative feedstocks, or bio-based options such as using natural fibers. However, a lack of good interfacial adhesion between these fibers and matrix and their poor resistance towards moisture limit their usage in overmolding. In addition, the interdiffusion process between matrix and fibers in prepregs and tapes and interphase formation between the insert and overmolded materials are still open innovative solutions to initiate the market growing. To conclude, overmolding with significant economic benefits paves a way to manufacture high-performance composites and parts in a single-step process, especially for automotive and aerospace industries. When the design tools combine with this scalable manufacturing process with an ideal material selection, overmolded parts having complex geometries with excellent bonding of dissimilar material systems can be achievable to meet the current market demands.

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.

Acknowledgments

This research was supported by TUBITAK 2244 - Industrial Ph.D. Fellowship Programme [Grant no: 118C055] funded by the Scientific and Technological Research Council of Turkey (TUBITAK), Farplas Automotive and Sabanci University.

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