# PREPERATION AND MECHANICAL PROPERTIES OF WOVEN FABRIC REINFORCED ALL POLYPROPYLENE COMPOSITES

# DOKUMA KUMAŞ TAKVİYELİ POLİPROPİLEN KOMPOZİTLERİN HAZIRLANMASI VE MEKANİK ÖZELLİKLERİ

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## ABSTRACT

In the present work, all polypropylene composites were prepared by film stacking technique using terpolymer and copolymer matrixes and woven fabric reinforcement. Composite sheets were prepared by use of hot press at different processing temperatures of 5, 10 and 15 °C over the melting temperature (Tm) of the matrixes in constant pressure of 8 MPa. Consolidation quality of composites was studied by scanning electron microscopy (SEM) and density measurements. Mechanical properties of composite sheets were subjected to tensile, flexural, and impact tests. The results showed that strength, stiffness and consolidation quality of composites increased by increasing the processing temperature while impact properties decreased. It was established that composite with terpolymer matrix demonstrated good strength, stiffness as well as consolidation properties in lower preparing temperature that indicate terpolymer matrix can expand the processing window in manufacturing of all polypropylene composites.

**Key Words:** Woven fabric, All polypropylene composite, Mechanical properties, Consolidation, Film stacking technique.

## ÖZET

Bu çalışmada, bütün polipropilen kompozitler, terpolimer ve kopolimer matrisleri ve dokuma kumaş takviyesi kullanılarak film stacking tekniği ile hazırlanmıştır. Kompozit levhalar, 8 MPa sabit basınç altında, sıcak pres kullanılarak, matrislerin erime sıcaklıklarının (Tm) 5, 10 ve 15°C üzerinde değişik işlem sıcaklıklarında hazırlanmıştır. Kompozitlerin haddeleme kalitesi, taramalı elektron mikroskobu (SEM) ve yoğunluk ölçümleri ile çalışılmıştır. Kompozit levhaların mekanik özellikleri çekme, eğilme ve darbe testlerine tabi tutulmuştur. Sonuçlar, işlem sıcaklığının artması ile kompozitlerin dayanımı, sertliği ve haddeleme kalitesinin arttığını ve darbe özelliklerinin azaldığını göstermiştir. Terpolimer matrisli kompozitler, düşük sıcaklıklarda iyi mukavemet ve sertliğin yanı sıra iyi haddeleme özellikleri de göstermişlerdir. Bu da, terpolimer matrislerin bütün polipropilen kompozitlerin üretiminde kullanılabileceğini göstermektedir.

**Anahtar Kelimeler:** Dokuma kumaş, Polipropilen kompozit, Mekanik özellikler, Haddeleme, Film stacking tekniği.

## 1. INTRODUCTION

In recent years, barriers and technical difficulties to recycling in traditional heterogeneous composites to comply with waste management regulations and environmental protection policies have drawn a great deal of research attention to find more convenient alternative composites. Polypropylene, as being one of the most widely used polymer in composite manufacturing, has gained more attention of researchers for the development of recyclable composites. Self reinforced polypropylene composites that are also called all polypropylene (PP) composites, are composites that both the reinforcing and the matrix materials are polypropylene, so they can be easily recycled after their useful life time. Due to identical material for matrix and reinforcing sections, setting a suitable range of preparing temperature that is called processing window, is the most challenging factor in composite manufacturing. Preparing

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of self reinforced all PP composites, have been introduced in different methods of composite processing such as hot compaction (1-7), consolidation of the co-extruded PP tapes (8-14), and film stacking methods (15-19). Hot compaction technique was developed by Hine et al., (1). In this method, small area of the oriented phase surface is melted and will be re-crystallized to form matrix phase. Besides the advantages including, good mechanical properties, and high quality single composite, this method requires precise control of the process system, due to its narrow processing window. In the next preparation technique; so called the consolidation method, co extruded tapes with PP copolymer, produce a shell and core structure tapes that benefits from larger processing window (8). However, production of co extruded tapes has restricted this method to constant reinforcing material. In the last attempt to extend processing window, film stacking technique was developed by Barany et al. (18). In this method, film layers act as the composite matrix which have lower melting point than reinforcing material and they will be laminated with reinforcement and hot compression will mold them at temperature higher than melting temperature (Tm) of the matrix. Film stacking method was regarded as a promising method in which the possibility of selecting matrix materials with lower melting temperature was a perceptible advantage. Today, the versatility of matrix materials like PP copolymer matrix or modification on homopolymer or copolymer matrix (20-23) and also reinforcement materials like textile fabric (24) ,carded mat (25) and non-woven's (17,26-27) are used in composite construction.

In recent years, extending the processing window has been one of the main concerns in production of all PP composites. Furthermore, from industrial point of view, it is easier to control the manufacturing conditions when the melting points of composite components are noticeable different. In order to improve the processing window in manufacturing of all PP composite, exploring new materials beside the mentioned innovated matrix modification in film stacking technique, will open a new approach in manufacturing of all PP composite. The aims of this study is to prepare all PP composite with woven fabric reinforcement and apply terpolymer matrix that has the potential of expanding the processing window, in manufacturing of woven fabric reinforced all PP composite by film stacking technique and investigate its mechanical performance in comparison with co polymer matrix.

## 2. MATERIAL AND METHOD

### 2.1. Materials and Their Properties

A plain woven tape fabric with a nominal weight of 180 g/m$^2$ (supplied from Pinak Company, Rasht, Iran) was used as reinforcing part of composite. Based on measurement of single tape of fabric, tensile strength and modulus was 280±20 and 2600±50 MPa respectively. Two different kinds of PP grades, poly ( propylene 95% wt /ethylene 5% wt ) random copolymer, (RP340N, Jam polypropylene Company, Assaloye, Iran) with melt flow index of 6 g /10 min at 230 ˚C and 2.16 kg load and poly (propylene 90 % wt /ethylene 5 % wt /butylenes 5 % wt ) random terpolymer (TD215 BF, Borseal™, Austria) with melt flow index of 6 g /10 min at 230 ˚C and 2.16 kg load was selected as matrix materials. To characterize melting point of PP yarn and matrix materials, differential scanning calorimetric (DSC) was used. DSC curves were recorded using DSC - 822 Mettler-Toledo instrument (Greifensee, Switzerland). 4-5 mg samples dried prior to measurements and the following temperature scans was used: Heating at the rate of 10 ˚C min$^{-1}$ to 210 ˚C.

The scanned curves are presented in Figure.1. It can be seen that the difference between melting point of PP yarn and terpolymer matrix (36 ˚C) is two times higher than copolymer matrix (18 ˚C). Lower melting point of terpolymer matrix comparing to copolymer matrix, can provide wide processing window in manufacturing of all PP composite.

### 2.2. Composite Preparation

Before composite preparation, thin layers of film with the thickness of 180 µm from PP matrixes were manufactured by single screw extruder (Dr Collin Gmbh, type E, Ebersberg, Germany) and chilled roll cast film device downstream. The barrel temperature was set to 180–210°C with a screw speed of 40 rpm. In order to do composite preparation, woven fabrics with dimension of 16 * 16 cm2 were cut as reinforcement and 8 pieces of fabric laminated with 9 layer of film. The nominal woven fabric reinforcement was kept approximately 50% in weight for all composites. Three temperatures of 5, 10, and 15 ˚C over matrix melting point, selected as processing temperature for preparing the composites during the hot press process. The layers of composite materials were put into the mold and after preliminary heating, hot compressed for 8-10 min under the pressure of 8 MPa by using conventional manual hot press process. In the next step, the pressed composites were cooled rapidly by cooling water to room temperature in 5 min under the same pressure. The proposed method was used to prepare approximately 3 mm thickness composite sheets. For comparison purposes, sheets from terpolymer and copolymer matrixes were prepared by above mentioned method.

### 2.3. Composite Characterization

Static tensile tests were performed on dumbbell specimens, 6 mm width and 33 mm length of narrow section, using Universal Testing Machine, Zwick/Roell Z050, according to ASTM D 638 with gauge length of 25mm and crosshead speed of 30 mm/min. Flexural 3 point bending test was carried out by Universal Testing Machine Zwick/Roell Z050 on 12*60 mm samples according to ASTM D790 with span length of 48 mm and 2 mm/min speed. Izod impact tests were performed on 12.7*6.35 mm samples according to ASTM D256 with 2.75 potential energy and 3.46 m/s speed. All mechanical tests were carried out on five samples for each test at 22 ˚C and 60 % RH laboratory conditions.

Composites consolidation quality was studied by scanning electron microscopy on cut surface of the composites. The samples were coated with a thin layer of gold prior to SEM investigation. The densities of composites were measured in pure ethanol at 23 ˚C according to EN ISO 1183-1.

## 3. RESULTS AND DISCUSSION

### 3.1. Consolidation Quality

The structure of composite is composed of layers of fabrics that bonded by matrix materials. Adhesion of the layers is one of the important factors that affect mechanical properties. There are many potential sources of void formation during the melting and flow of matrix into the woven fabric [16]. Plain pattern of the woven fabric produces high crimp angle in tape yarn and causes the interlace contact in fabric that reduces the uniformity of the fabric surface. This results in void formation during preparation of composite.

In order to inspect the consolidation quality and void content of composites, scanning electron microscopy (SEM) was used. Figure.2 shows cross sections of composites with terpolymer and copolymer matrixes at different processing temperatures. The void defects are shown with arrows in pictures. We can observe that the void defects almost appear in contact areas of PP yarns with matrix. It is deducted that the quantity and size of the void defects would be decreased with increase of preparation temperature and hence higher fiber to matrix adhesion will be happened. The possible description is that at higher temperature processing, the viscosity of the matrix reduces and the flow of the matrix can better impregnate the surface of the fabric and good matrix to fiber adhesion could be occurred. The consolidation quality of composites also was confirmed by density measurements.

Table 1 tabulated the density of composites for different processing temperatures. It was observed that the density of composites increased by increasing the composite preparation temperature as a result of lower void defects in the composite structure. Composites with terpolymer matrix demonstrated similar density values in comparison with copolymer matrix that shows good consolidation quality and interfacial adhesion are happened at lower processing temperatures. This result is also confirmed by SEM pictures. It can be seen from table. 1 that the density of composites with both copolymer and terpolymer matrixes are slightly higher than those of PP tapes (0.851 g/cm3). This result is compatible with reports of Alcock et al (9) and Abraham (21) that studied on co extruded PP tape composites. The possible reason behind is that during the manufacturing of PP tape yarn, micro void appears and thus reduces the density of PP yarns. Preparing of composite under high pressure and temperature, can reduce the micro voids which result in higher density values.

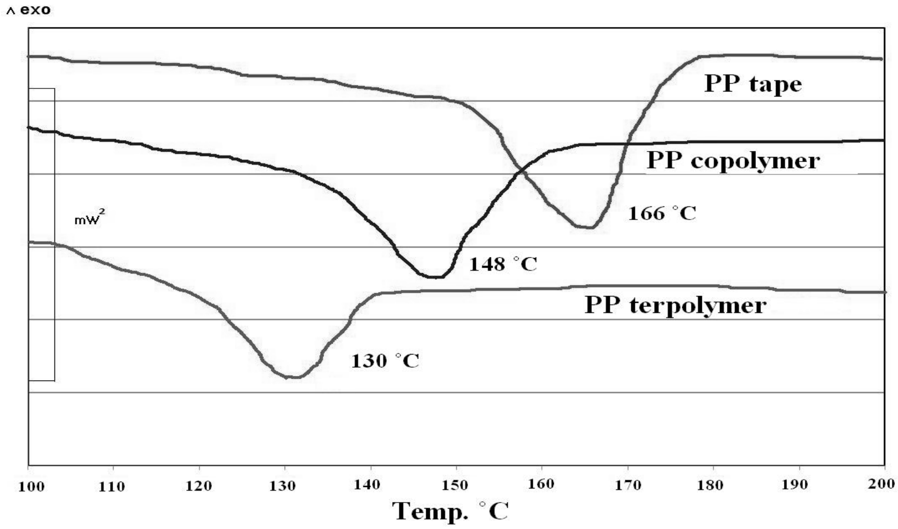

**Figure 1.** DSC curves of composite components (2nd heating run)

**Table 1.** Density of PP tape, matrixes and composites at processing temperature of +5, +10 and +15 °C over melting point of related matrix

| Materials | Processing Temperature over matrix melting point [° C] | Density[ g/cm3] |
|---|---|---|
| Composite with copolymer matrix | +5 | 0.852 ±0.002 |
| | +10 | 0.888 ±0.002 |
| | +15 | 0.902 ±0.002 |
| Composite with terpolymer matrix | +5 | 0.850 ±0.002 |
| | +10 | 0.887 ±0.002 |
| | +15 | 0.899 ±0.002 |
| Copolymer matrix | - | 0.896 ±0.001 |
| Terpolymer Matrix | - | 0.901 ±0.001 |
| PP tape | - | 0.851 ±0.001 |

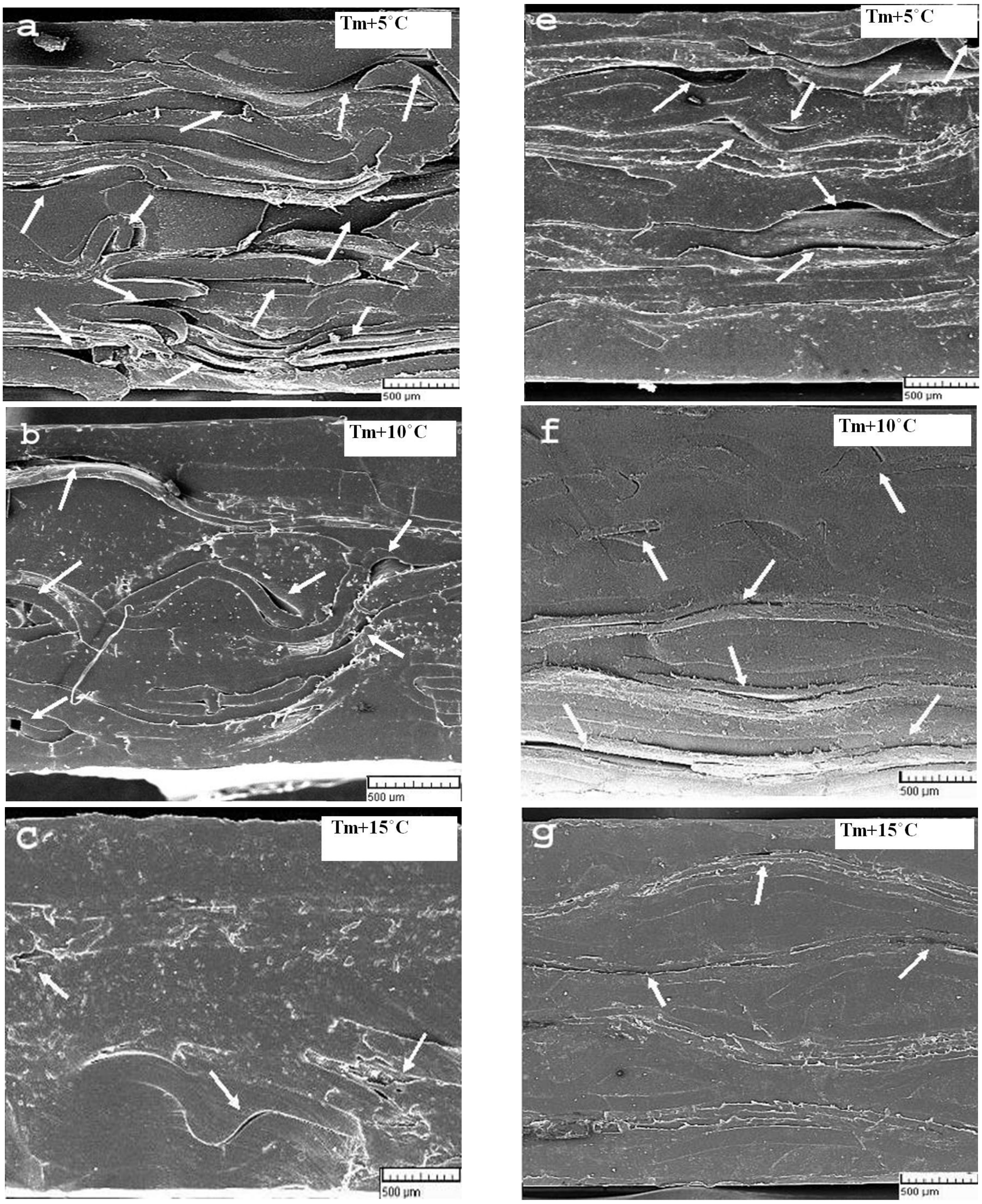

**Figure 2.** Scanning electron microscopy of cut cross section of composites at different processing temperature: a, b, c composite with Terpolymer matrix and e, f, g composite with Copolymer matrix

### 3.2 Static Tensile Test Results

Figure. 3 display the tensile strength and modulus for the matrix materials and related composites. It is obvious that both tensile stress and tensile modulus of composites increased with increasing consolidation quality by composite preparing temperature. In composite with copolymer matrix at processing temperature of Tm+15˚C, mechanical properties decreased. The possible explanation is that the processing temperature of this composite is near to melting point of PP yarn so the partial melting and molecular relaxation in PP yarn could be happened. From Figure.3, it can be seen that the maximum value in tensile properties of terpolymer composite can be achieved at lower composite preparing temperature (Tm+15˚C= 140˚C) in comparison with copolymer matrix (Tm+10˚C= 158˚C). However composite with copolymer matrix, exhibited better tensile properties at different processing temperature due to its better consolidation quality. Figure. 4 demonstrate a typical failure behavior of tensile test of composites at different processing temperatures. It can be seen that the tensile failure mode of the composites changes by increasing the processing temperature and consolidation quality. At low processing temperature where the consolidation degree is poor, the failure occurs with tape pull out and fiber breakage. As the processing temperature increases and consolidation degree improves, the composites breaks in normal way with less fiber pull out.

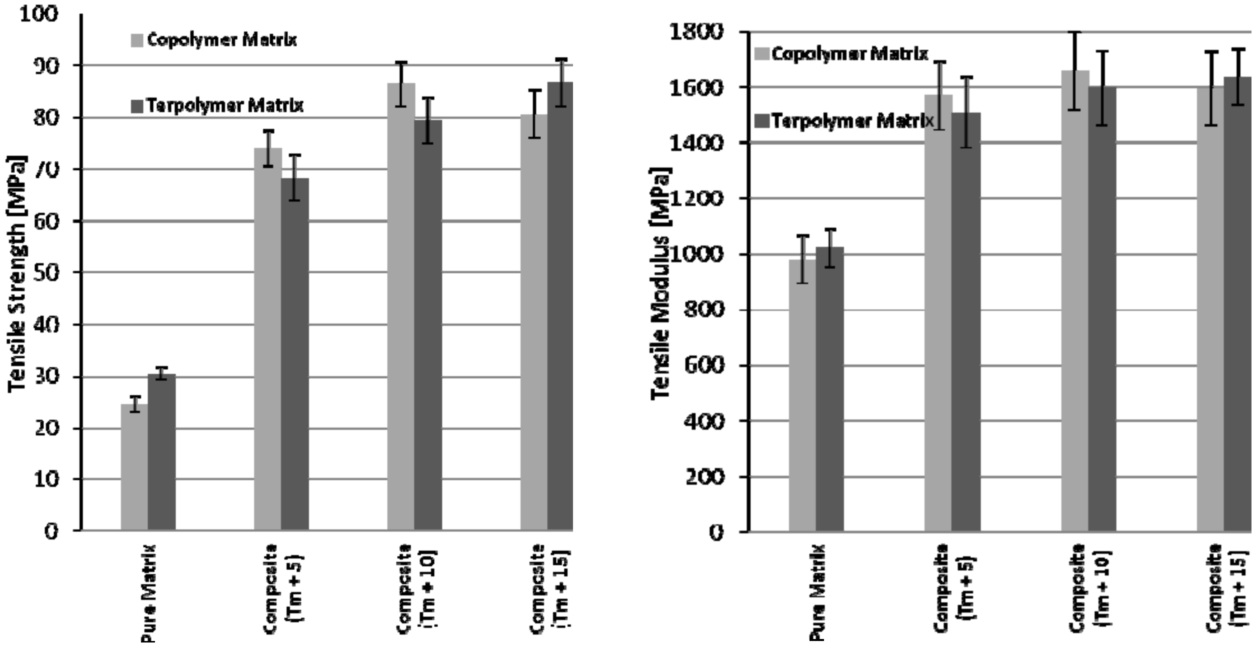

**Figure 3.** Tensile strength and tensile modulus for pure matrixes and composites at processing temperature of 5, 10 and 15 ˚C over melting point of related matrix

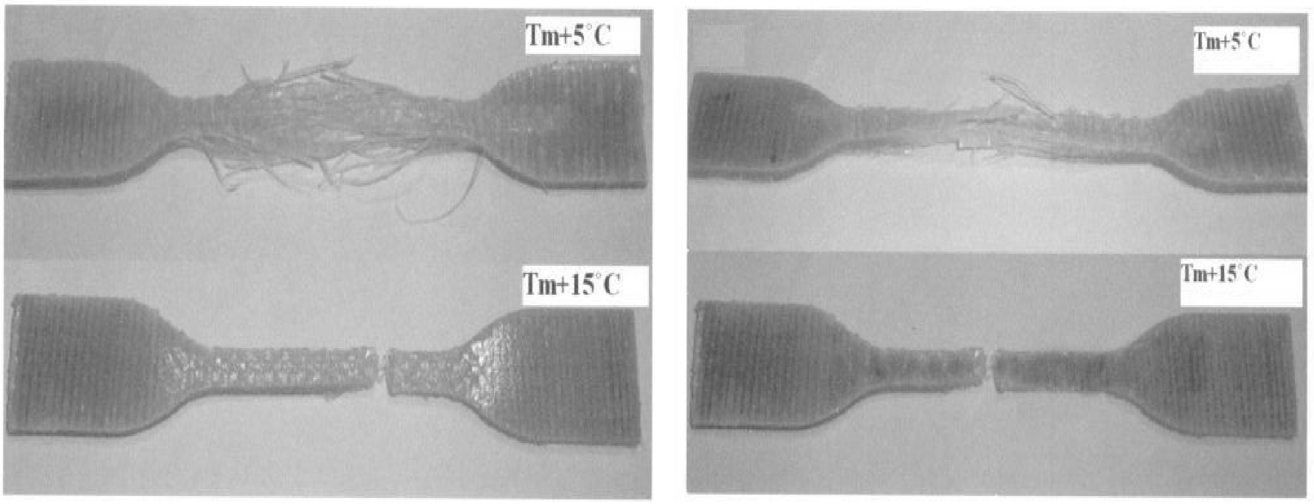

**Figure 4.** Typical failure behavior of tensile specimens of composites at different processing temperature. Left: terpolymer matrix and right: Copolymer matrix

### 3.3. Flexural Test Results

Figure.5 shows typical flexural stress – strain curve of composites with terpolymer and copolymer matrixes at processing temperature of Tm+15 ˚C. It is obvious that there is double yielding behavior in curves. Double yielding behavior in flexural stress-strain curve of all PP composite with coextruded tapes has been reported by Banik (28). It seems that the mechanism of double yielding is related to different structures of matrix as well as reinforcing parts inside the composite. It is suggested that the first yielding point can be responsible for matrix material and the second yielding point can be responsible for reinforcing woven fabric. Figure.6 depicts the flexural strength and modulus against processing temperature. It is obvious that flexural stress and modulus increased by higher processing temperature. From Figure.6, we can conclude that the similar behavior that found for tensile test results can be extended to flexural test results.

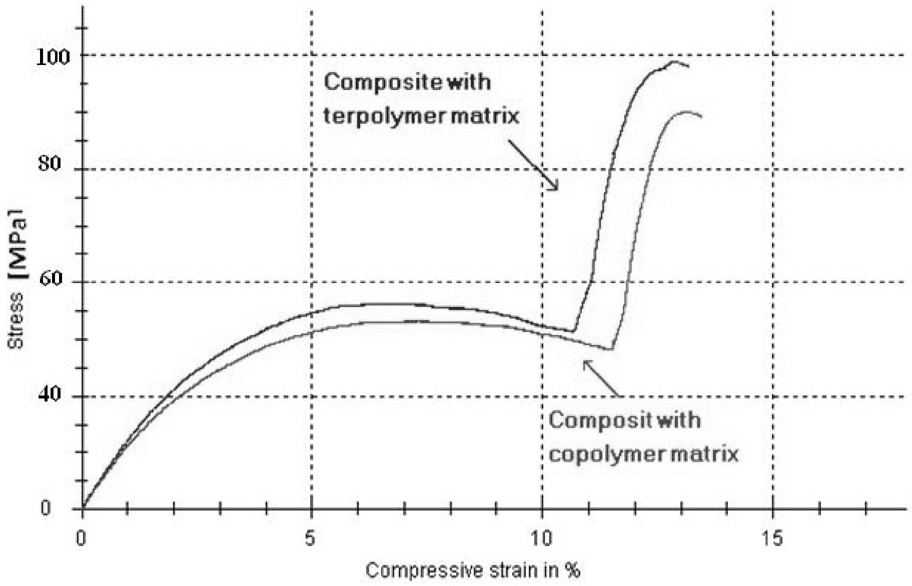

**Figure 5**. Typical flexural stress -strain curves of composites with terpolymer and copolymer matrixes at Tm+15˚C processing temperature

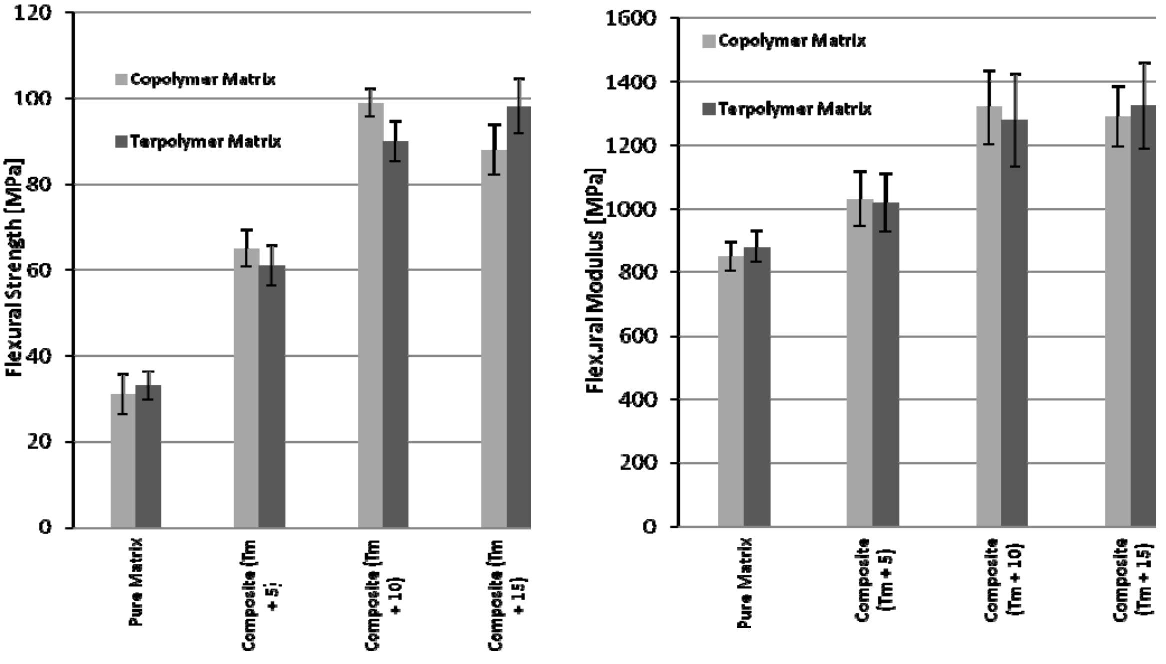

**Figure 6**. Flexural strength and flexural modulus for pure matrixes and composites at processing temperature of 5, 10 and 15 ˚C over melting point of related matrix

### 3.4 Impact Test Results

Figure.7 shows typical energy, force against time curve of impact test of composites with terpolymer and copolymer matrixes at the processing temperature of Tm +5˚C and related matrix materials. It is clearly recognized that the composites can absorb energy more than five times comparing to related matrixes.

The mechanism of energy absorption in woven fabric all PP composites is related to delaminating of the fabric layers and tapes pull out during the test. This means that poor consolidation result in better energy absorption while it can lead to poor tensile and flexural properties.

Figure.8 demonstrated the effect of processing temperature on impact strength of composites. In both composites with terpolymer and copolymer matrixes, the impact strength has a reverse behavior with increasing the processing temperature. Also the absorbed energy of composite with terpolymer matrix is higher than with copolymer matrix that is due to their different consolidation quality.

Figure.9 demonstrates the typical failure behavior of Izod impact specimens at processing temperature of Tm+5 ˚C and Tm+15 ˚C. When the consolidation is poor, the failure occurs typically by delimitation and tapes pull out. With increasing consolidation quality, the delaminating becomes less pronounced when the specimens break.

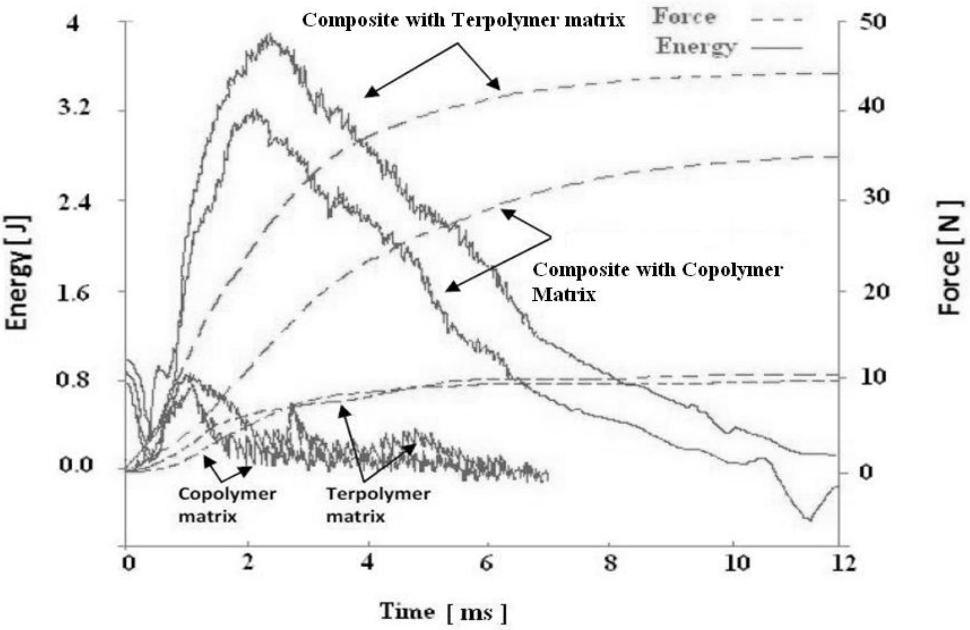

**Figure 7.** Typical force,energy vs. time curves recorded during Izod tests for pure matrixes and composites with terpolymer and copolymer matrixes.( processing temperature Tm+5˚C)

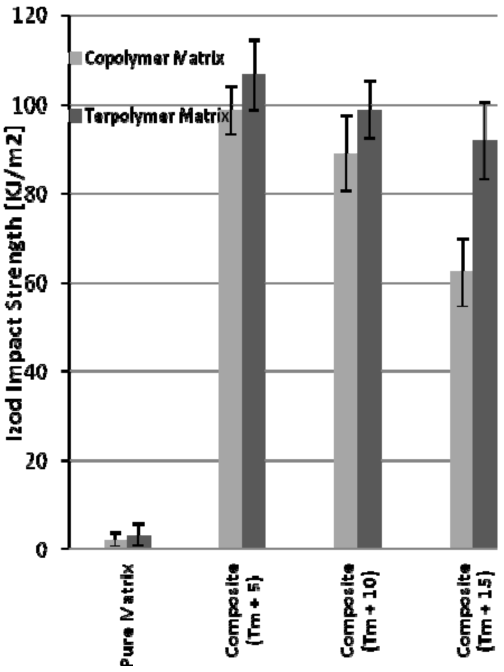

**Figure 8.** Izod impact strength for pure matrixes and composites at processing temperature of 5, 10 and 15 ˚C over melting point of related matrix

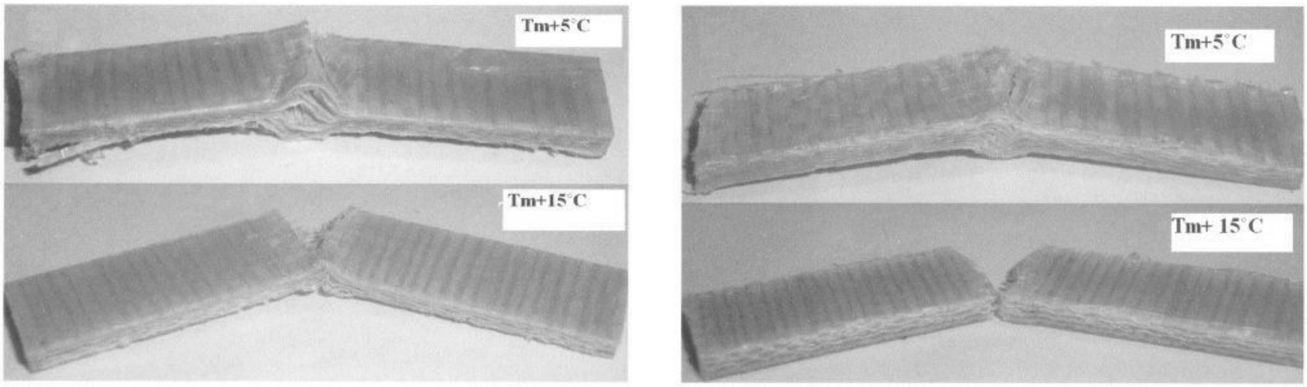

**Figure.9** Typical failure behavior of Izod impact specimens of composites at different processing temperature. Left: terpolymer matrix and right: Copolymer matrix

## 4. CONCLUSIONS

In the present work, woven fabric reinforced all polypropylene composites were successfully prepared by film stacking technique and applying poly (propylene/ ethylene/butylene) random terpolymer and poly (propylene/ethylene) random copolymer as matrix materials. By increasing the preparing temperature of composite, consolidation degree, tensile and flexural properties showed better properties both in composite with copolymer and terpolymer matrixes, whereas the impact properties showed a reverse behavior. The maximum values of tensile and flexural properties can be achieved in lower composite preparing temperature by applying terpolymer matrix.

Based on the current work, poly (propylene/ethylene/butylene) random terpolymer matrix can be applied successfully in manufacturing of woven fabric reinforced all PP composites with reasonable mechanical properties in order to expand the processing window.

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