

## Effect of Adding Zirconia and Alumina Nanoparticles on the Tensile and Flexural Properties of Polymer Blend Based on Polypropylene

Teeb Adnan Mohameed<sup>1\*</sup>, Sihama Issa Salih<sup>2</sup>, Wafaa Mahdi Salih<sup>2</sup>

<sup>1</sup>. Central Organization for Standardization and Quality Control Baghdad, Iraq.

<sup>2</sup>. Materials Engineering Department, University of Technology / Baghdad-Iraq.

\*Corresponding Author: Teeb Adnan Mohameed

### Abstract

Polypropylene is an important material used in many structural applications, including automotive and aircraft parts as well as machine parts. In this paper, the properties of polypropylene were improved by blending it with 5% ratio of poly methyl methacrylate (PMMA) and adding nanoparticles powders of zirconia ( $ZrO_2$ ) and alumina ( $Al_2O_3$ ) in different ratios (0.3, 0.5 and 0.7%) to polymer blend, which consists of [PP: 5% PMMA and adding 1% of (polypropylene-grafted-malic anhydrated (PP-g-MA) as compatibilizing agent)]. Polymer blend and its nanocomposites were prepared by melt blending technique using twin screw extruder. Investigation of some mechanical properties and the morphology structure test of the nanocomposites samples was studied. The results illustrated that the mechanical properties increased when adding nanoparticle ( $ZrO_2$  and  $Al_2O_3$ ) to polymer blend (PP: 5%PMMA: 1%(PP-g-MA)) and the highest values were obtained for tensile strength, modulus of elasticity, flexural strength and flexural modulus, at 0.3% ratio of ( $ZrO_2$  and  $Al_2O_3$ ) nanoparticles in the blend. The fracture surface morphology showed that the addition of nanoparticles led to reduce the micro pores that exist in the morphology structure of polymer blend.

**Keywords:** *Polymer blend, Nanocomposite, PP, PMMA.*

### Introduction

The characteristics of the composite materials depend on the properties for both the matrix and reinforcing materials, the interface among their processing conditions, wettability between their components and component melt viscosities [1]. The reinforcing materials are used to improve and increase the strength and stiffness of the matrix materials, thus the matrix and reinforcing materials should have a strong bond to perform the basic function [2, 3].

The nanocomposite materials are more important from the categories of advanced materials due to have unique properties such as high strength, thermal, gas-barrier, and flame resistance and others, however these enhanced the characteristics by the incorporation of nanofillers with preferably less than 100 nm in size [4]. Recently, the polymeric materials are used in industry, medicine and structural application but

because they have relatively low properties, their use in these applications is limited. Thus, the used many methods to increase the polymers properties are mixing the different polymeric materials, using co-polymer with new backbone chain and also by using reinforcing materials with high strength [5]. In the last years, many researchers studied to improve the polymeric properties by the addition of more than one type of reinforced materials to the neat polymer as well as to the polymer blend to prepare the hybrid composites.

Jia-Horng, et al. examined the influence of the content of HDPE on the properties and compatibility of poly blends. The SEM, DSC and XRD, results showed that PP and HDPE are incompatible polymers with PP being a continuous phase and HDPE being a dispersed phase. The FTIR results revealed that the combination of HDPE does not

influence the chemical structure of PP, indicating that the poly blends are made of a physical blending. However, according to mechanical property test results, the combination of HDPE improves the impact strength of PP [6]. Another study focused on the influence of processing conditions on the mechanical properties of polypropylene (PP) and PP/mesoporous silica-hydroxyapatite (PP/MCM-41-HA) hybrid nano-composites. The mechanical properties of PP were enhanced by adding MCM-41-HA nanoparticles and maleic anhydride-grafted poly-propylene (PP-g-MA), which were prepared using the melt intercalation technique in an internal mixer.

The PP/MCM-41-HA hybrid nanocomposite demonstrated a sensible enhancement of mechanical properties over the neat PP [7]. Other researchers studied the influence of reinforcing LLDPE/PA12 blends by modified nanoparticles, particularly located at the interface, on the morphology and mechanical properties, and clay addition with ratio 1% led to an increase of Young's modulus. But the raised clay fractions lead to improve the clay interphase that stabilizes the morphology and prevents the contact between the two polymers, Young's modulus is quasi-constant [8].

The underlying mechanism of the mechanical properties improvement for composites (polypropylene/low-density polyethylene) with different contents of filler masterbatch (nano-silicon dioxide, compatibilizer, lubricant agent, and antioxidant agent) was investigated. The results indicated that the mechanical properties, including tensile strength, moduli, and elongation can be drastically improved with the addition of the filler masterbatch.

The results also suggested that the compatibility of the two phases increases with the increase in the filler masterbatch. Furthermore, it was also found that there is a close relationship between the mechanical properties and morphological structures, which are improved by the existence of the filler masterbatch [9].

Another research was about the hydrolyzed powder coating recyclers that were used as filler material in a polypropylene matrix. The tensile strength of compounds decreased with increasing amount of hydrolyzed powder coating wastes in PP.

This decrease in tensile strength was due to possible in- homogeneities of the filler distribution in the polymer matrix. However, the tensile strength improved by the addition of MAH as a compatibilizer between matrix and filler particles, and the tensile stresses of the samples increased compared to samples without MAH. No gaps between matrix and filler particles were detected in the cross sections of the MAH-reinforced [10].

The interaction between nylon 6 (PA6), polypropylene (PP) and poly (lactic acid) (PLA) was reported. To improve the compatibility between these immiscible polymers, a reactive compatibilization approach was used through extrusion with maleic anhydride grafted polypropylene (PP-g-MA).

To further improve the compatibility of the phases, the PP-g-MA grafting process was observed to have a dramatic effect on the compatibility of both the binary and ternary blends, while the a drastic reduction of the PP dispersed phases particle size was observed, the affinity between PLA and PP was also highlighted [11]. This article focuses on the effect of compatibilizer materials (PP-g-MA, POE-g-MA and EVA-g-MA) with the same functional group as well as their mixtures and recycled polyethylene terephthalate (r-PET) content on the mechanical properties of compatibilized r-PET/PP blends.

The results depicted that the introduction of r-PET into PP matrix increased the tensile and flexural strength of PP. Addition of PP-g-MA improved the tensile and flexural strength of r-PET/PP blends, and the introduction of POE-g-MA or EVA-g-MA increased the impact strength of r-PET/PP blends, this provides polymer blends (r-PET/PP) with high strength and toughness by compatibilization of the mixtures of PP-g-MA and POE-g-MA and EVA-g-MA [12]. The aim of this work is to improve the mechanical properties of the polymer blend (PP: 5%PMMA: 1 %( PP-g-MA)) that based on polypropylene by adding nanoparticles of zirconia and alumina in individually form to it, for used in structural application.

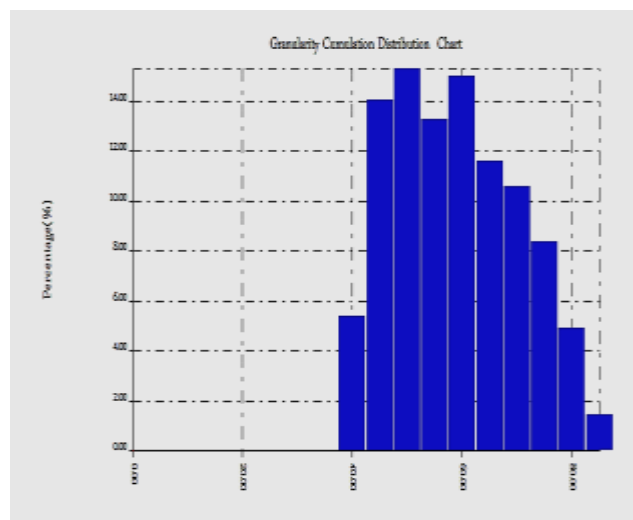
## Materials and Methods

### Materials

Materials used in this work are

polypropylene (PP) grade (500P for extrusion applications) provided from Sabic Company as a pellets form. Ultra-high molecular weight polyethylene (UHMWPE) was supplied from China for extrusion in a pellet form, with specific gravity ( $0.9 \text{ g/m}^3$ ) and melt flow rate ( $2 \text{ g/10 min}$ ) and polypropylene-grafted maleic anhydride (PP-g-MA) was provided from China in a powder form with melt temperature ( $150^\circ\text{C}$ ).

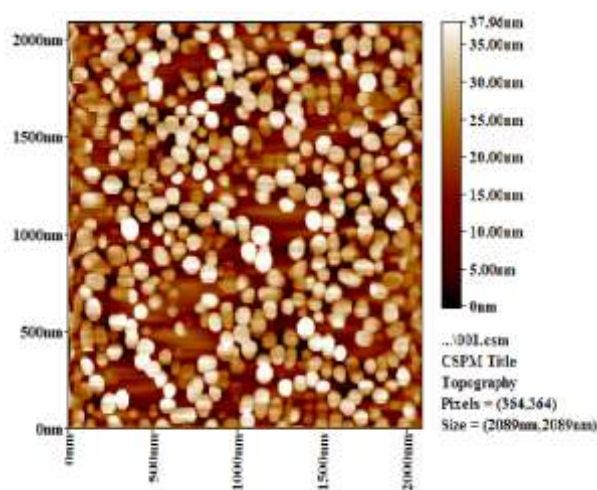
Zirconia ( $\text{ZrO}_2$ ) and Alumina ( $\text{Al}_2\text{O}_3$ ) nanoparticles were provided from China. Atomic force microscopy (AFM) test was used to measure the average particle size of nanoparticles. Figure (1) (a and b) and Figure (1) (c and d) depict the particles size distribution and image in three-dimensional (XYZ) by AFM for zirconia and alumina powder, respectively.



Avg. Diameter: 56.45 nm

$\leq 50\%$  Diameter: 55.00 nm

(a) Three-dimensional (XYZ) AFM pictures

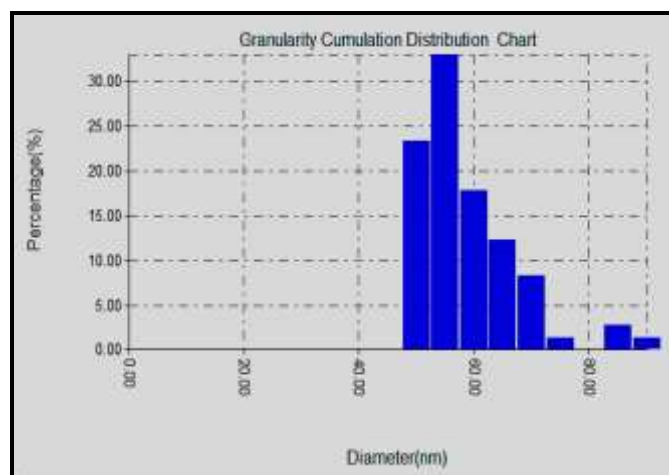


$\leq 10\%$  Diameter: 40.00 nm

$\leq 90\%$  Diameter: 70.00 nm

(b) Granularity accumulation distribution char.

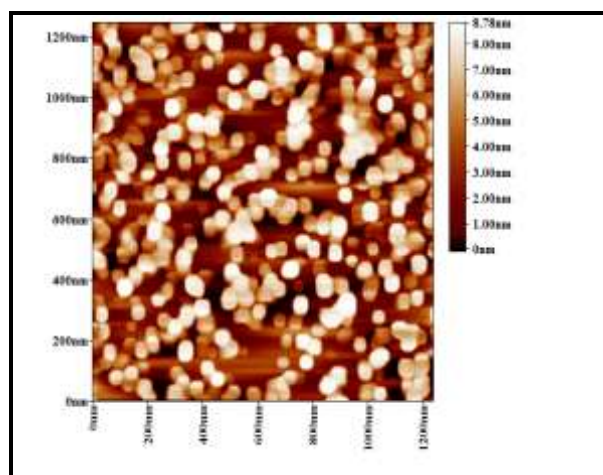
(A):  $\text{ZrO}_2$  nanoparticle.



Avg. Diameter: 56.29 nm

$\leq 50\%$  Diameter: 50.00 nm

(c) Three-dimensional (XYZ) AFM pictures



$\leq 10\%$  Diameter: 0 nm

$\leq 90\%$  Diameter: 65.00 nm

(d) Granularity accumulation distribution char

(b):  $\text{Al}_2\text{O}_3$  nanoparticle.

Figure 1: Atomic Force Microscopy of the nanoparticles where (a):  $\text{ZrO}_2$  and (b):  $\text{Al}_2\text{O}_3$

## Experimental Procedure

The nanocomposites of ternary polymer blend were prepared by melt blending technique using a twin - screw extruder. All components of the nanocomposites (pellets polymer, powder polymer and reinforcing

powders ( $\text{ZrO}_2$  and  $\text{Al}_2\text{O}_3$ ) nanoparticles), were putt inside oven at  $40^\circ\text{C}$  for 20 min to remove the moisture. The components of composites materials were weight according to that mentioned in Table (1), after that, each set of composite materials was melted in the twin - screw extruder machine with an

extrusion speed of 20 rpm at temperatures according to the three zones of extrusion (feed zone, compression zone and melting

zone) that are 195 °C, 205°C and 210 °C, respectively to obtain sheets with dimensions of (10 × 200 × 5 mm).

**Table 1: The weight percentages for components of composites materials**

The weight percentages of composites components	Weight Percentages of Reinforcing Powders (X%)		
	Sample 1	Sample 2	Sample 3
(94%PP: 5% PMMA: 1% (PP-g-MA)): X%ZrO <sub>2</sub>	0.3	0.5	0.7
(94%PP: 5% PMMA: 1% (PP-g-MA)): X%Al <sub>2</sub> O <sub>3</sub>	0.3	0.5	0.7

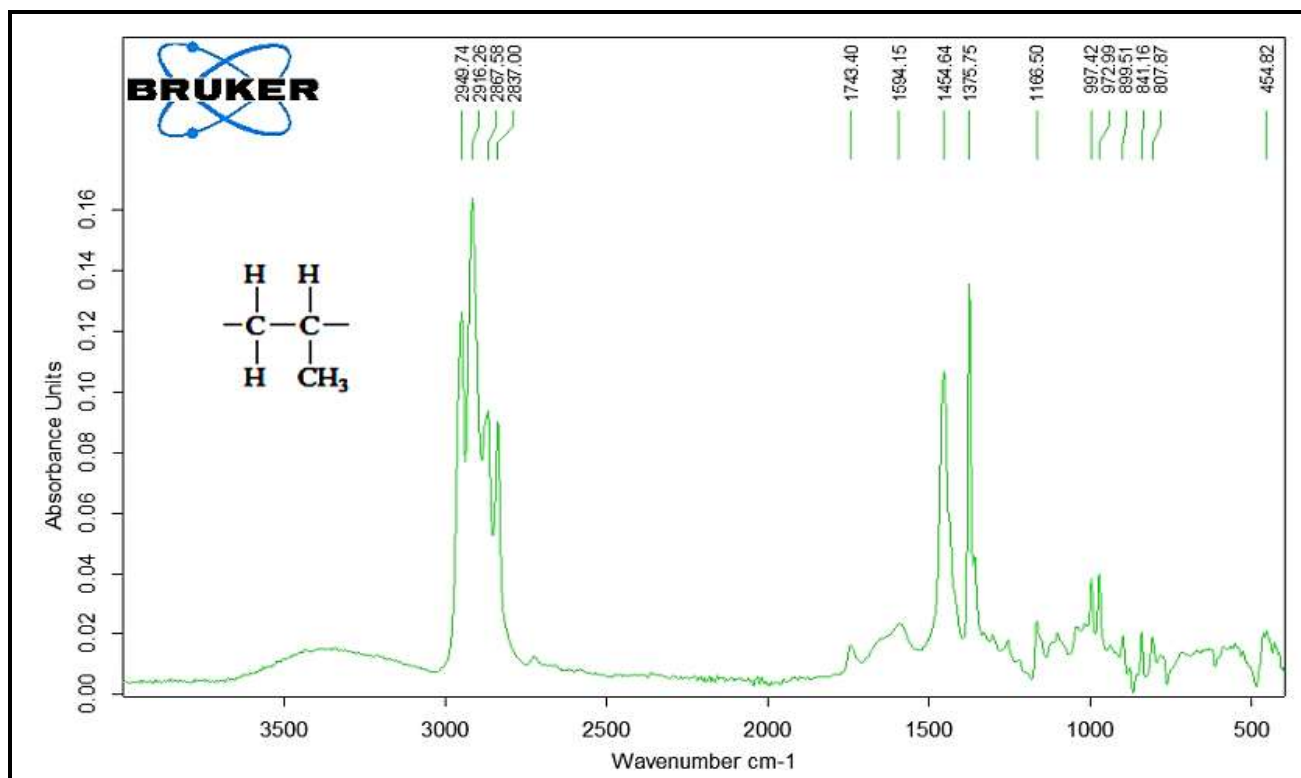
### FTIR Spectrum for Polymer Blend Nano-composites

The spectra of nanocomposites based on polymer blend (PP: 5%PMMA: 1 % (PP-g-MA)) which was reinforced with two types of inorganic nanoparticles (zirconia and alumina) added individually and at the different ratios (0, 0.3, 0.5, and 0.7%) were shown in Figures (3 and 4) respectively. From these figures it is observed that all the characteristics vibration bands of polypropylene in Figure (2) and of ternary polymer blend (PP: 5%PMMA 1% (PP-g-MA)), it has appeared in the FTIR spectra for their nanocomposites ((94%PP: 5%PMMA: 1% (PP-g-MA)): X% ZrO<sub>2</sub> or X%Al<sub>2</sub>O<sub>3</sub>), these spectra in a good agreement with that reported by [13 and 14].

Moreover, several peaks related (PP-g-MA) are observed in spectra of polymer blend and its composites. Furthermore, there are several peaks related to zirconia nanoparticle are observed in spectra of nanocomposites that reinforced by zirconia nanoparticles

(Figure (3), the peak located at (557.65) cm<sup>-1</sup> for vibration of (Zr–O) bond and another peak located at (470.01) cm<sup>-1</sup> of vibration of (O–Zr–O) bond [15], while the peak at 752.70 cm<sup>-1</sup> is attributed to Zr–O stretching vibrations at ZrO<sub>2</sub> nanoparticles [15 and 16]. The presence of Al<sub>2</sub>O<sub>3</sub> is also detected in spectra of nanocomposites that reinforced by alumina nanoparticles (Figure (4)).The main peaks at 457.79, 591. 51, and 601.02 cm<sup>-1</sup> can be assigned to the Al–O stretching mode in octahedral structure; [17, 18 and 19].

Moreover, from these infrared spectra, it was observed there is no shifting in peaks of characteristic frequencies of nanocomposites, relative to those spectra of polymer blend. Except, there is a clear height in the intensity of peaks with addition of zirconia and alumina nanoparticles, in the blend. This indicates to occurrence of physical bonding and absence from any chemical reactions or cross linking that may be occur as a result of the blending process between all components of the nanocomposite materials [5].



**Figure 2: FTIR spectrum for neat polypropylene material**



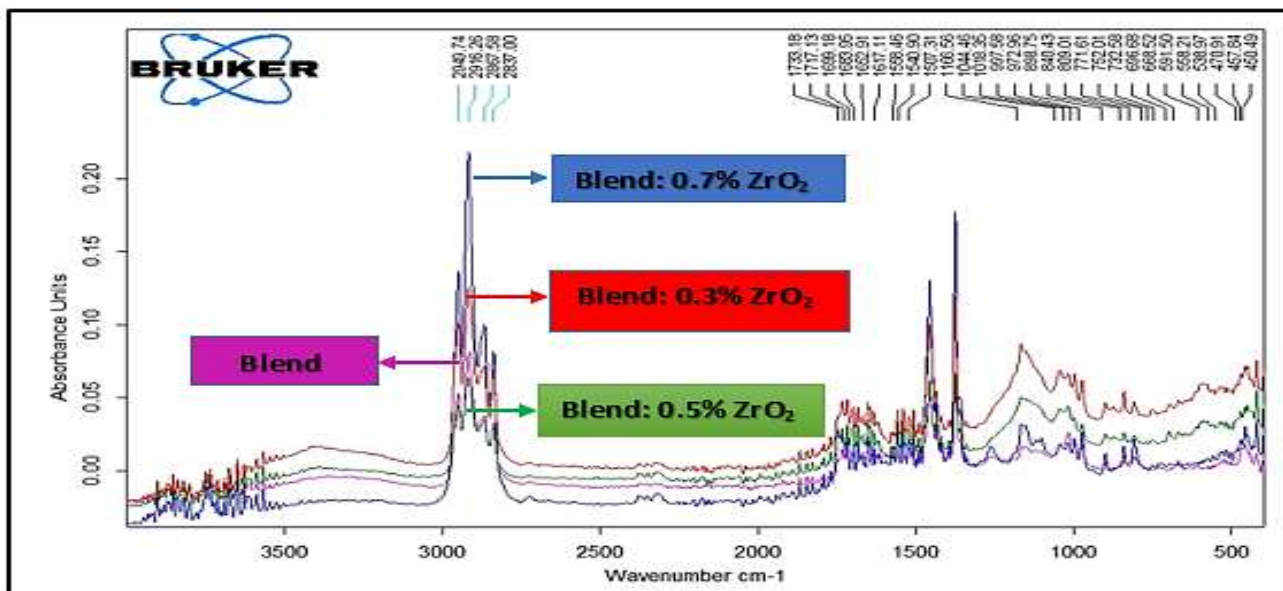


Figure 3: FTIR spectra for polymeric blends (PP: 5% PMMA: 1% (PP-g-MA)) and its nanocomposites as a function of  $ZrO_2$  nanoparticles content in blend

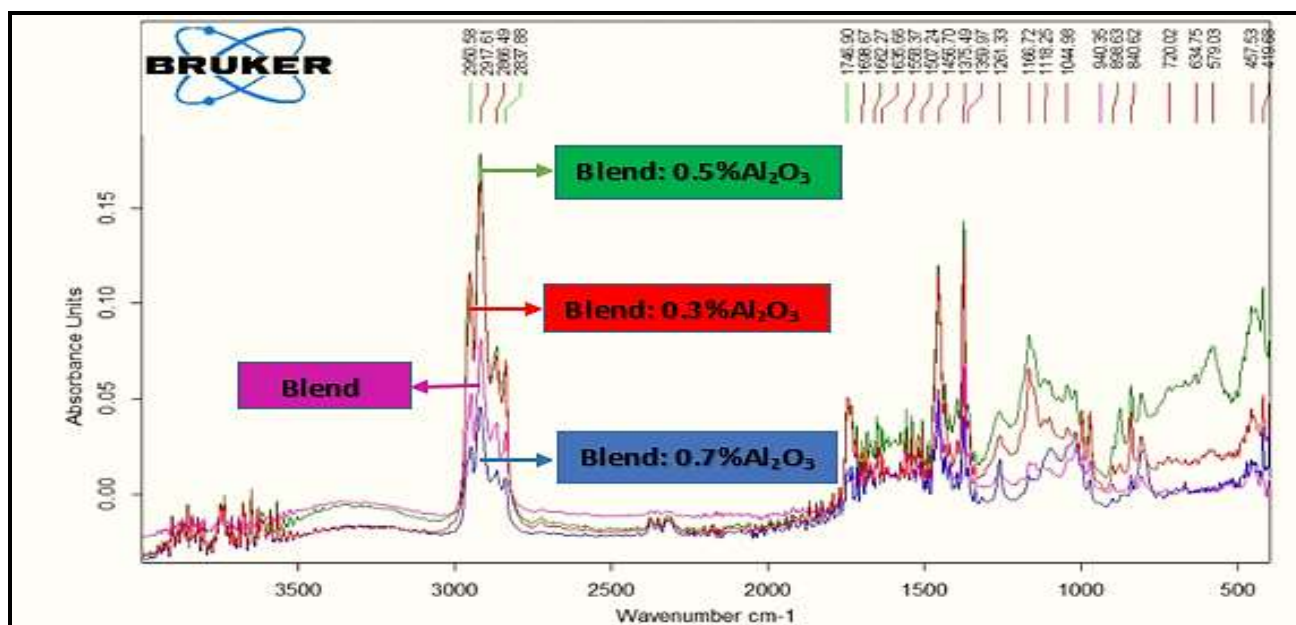


Figure 4: FTIR spectra for polymeric blends (PP: 5% PMMA: 1% (PP-g-MA)) and its nanocomposites as a function of  $Al_2O_3$  nanoparticles content in blend

## Mechanical Tests

The tensile test was done according to ASTM standard D638-87 [20]. By using machine (model WDW 200 E), the test was conducted with speed of cross-head (5 mm/min). Flexural test was conducted by using a three-point method according ASTM D790 standard [21], by a test machine, model (WDW 200 E), the test was carried out with speed of cross-head 2 mm/min, all the test were performed at a room temperature ( $23 \pm 5^\circ C$ ) and atmospheric pressure.

## Tensile Test Results

Tensile inspection was mainly achieved to investigate the (stress-strain) behavior between two groups of nanocomposites that

based on polymer blends which are ((PP: 5%PMMA:1%(PP-g-MA)):X% $ZrO_2$ ) and ((PP:5%PMMA:1%(PP-g-MA)): X% $Al_2O_3$ ) that are shown in Figures (5) and (6), respectively. It is noted that there is no significant difference in the behavior of the (stress stress) curves for both groups of nanocomposites, where it was observed that this behavior changes from a hard and tough behavior of the polymer blend material to a hard and strong behavior when the addition of zirconia and alumina nanoparticles to it. This can be attributed to the nature of the mechanical properties of both zirconia and alumina, both of which have a high rigidity compared to the components of the polymeric blend which have a flexible property [22].

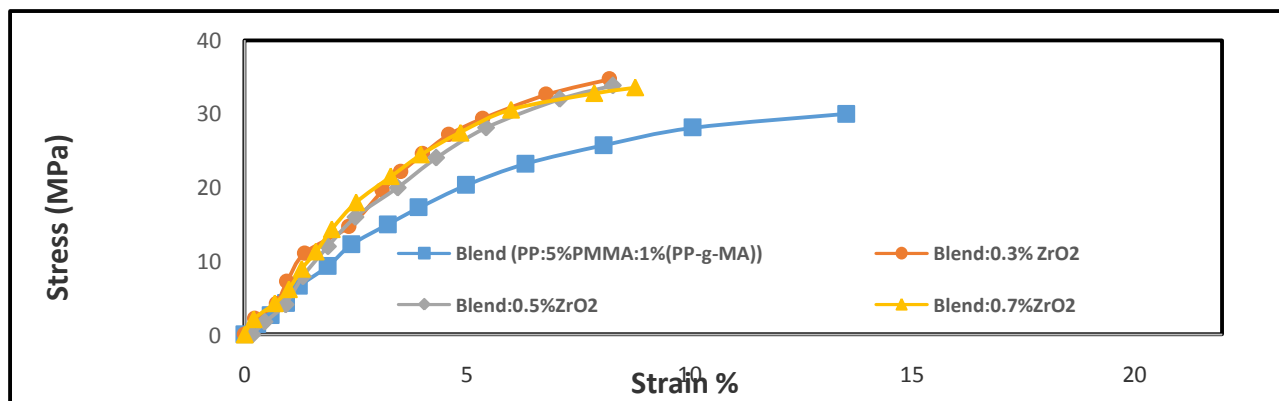


Figure 5: Stress-strain behavior of ((PP: 5%PMMA: 1% (PP-g-MA)): X%ZrO<sub>2</sub>) polymer blends nanocomposites

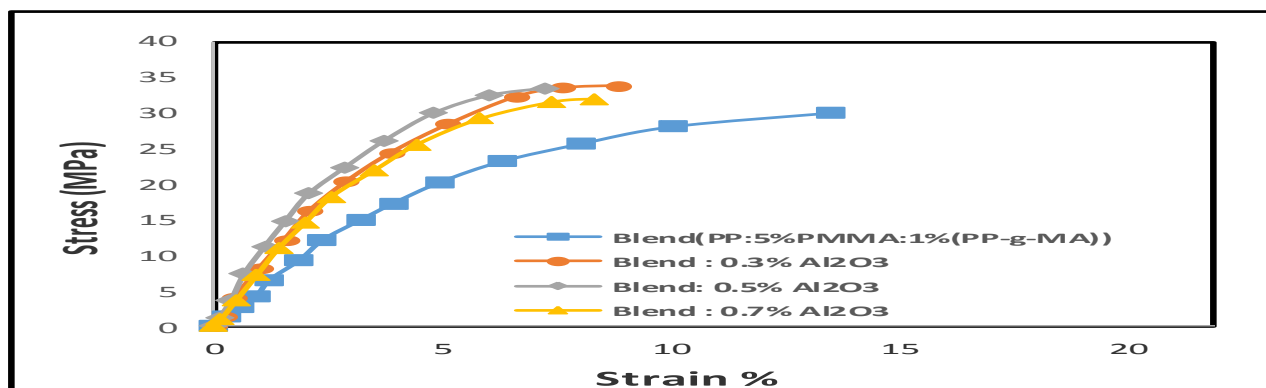


Figure 6: Stress-strain behavior of ((PP: 5%PMMA: 1% (PP-g-MA)): %Al<sub>2</sub>O<sub>3</sub>) polymer blends Nanocomposites

Figures (7) and (8) elucidate the effect of adding zirconia and alumina nanoparticle on the tensile strength and modulus of elasticity for nanocomposites, respectively. From these figures, it was noticed that the addition the nanoparticles increased the values of tensile strength and modulus of elasticity and arrived to the highest values at 0.3% wt. For each of the zirconia and alumina nanoparticles content in composite, the maximum values of tensile strength reached to 34.72 MPa and 33.9 MPa, respectively. These samples withstand the maximum load as compared to samples that contain the higher ratios of nanoparticles, where increasing the content ratios of zirconia and

alumina to higher than 0.3% led to decrease these values, but they remained higher than the base material of the polymeric blend (PP: 5%PMMA: 1%(PP-G-MA)). On the other hand, the nanocomposites samples ((PP: 5%PMMA: 1%(PP-g-MA)): X%ZrO<sub>2</sub>) that contain zirconia nanoparticles have the highest values of tensile strength as compared with their counterparts of the other group samples that contain alumina nanoparticle ((PP: 5%PMMA: 1%(PP-g-MA)): X%Al<sub>2</sub>O<sub>3</sub>). This is related to the nature of both zirconia and alumina nanoparticles and depended on the compatibility range between nanoparticles as reinforcement material and the components of polymer blend as matrix material [23, 24].

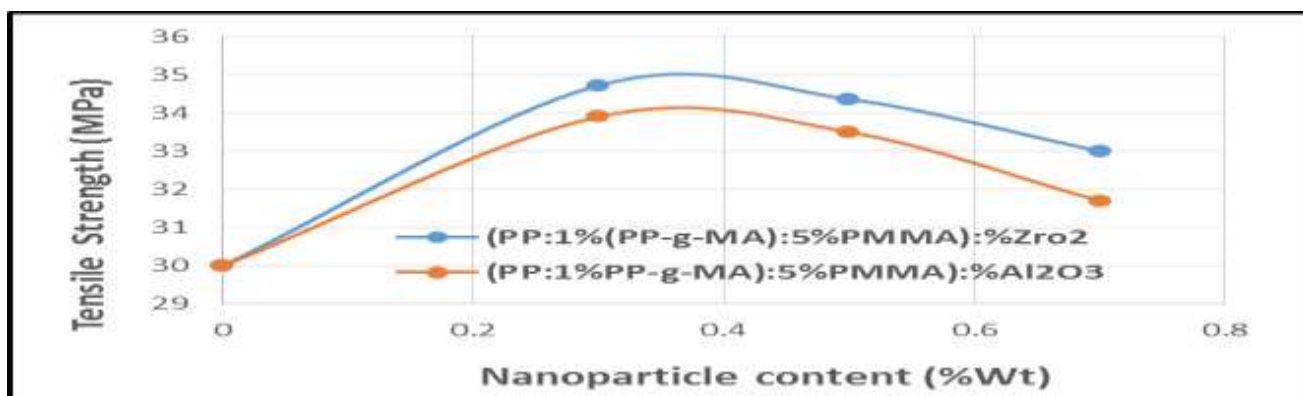


Figure 7: Tensile strength for Nanocomposites samples ((PP: 5%PMMA: 1 % ( PP-g-MA)): X%ZrO<sub>2</sub> or X%Al<sub>2</sub>O<sub>3</sub>) as a function of Nanoparticle content in composite.

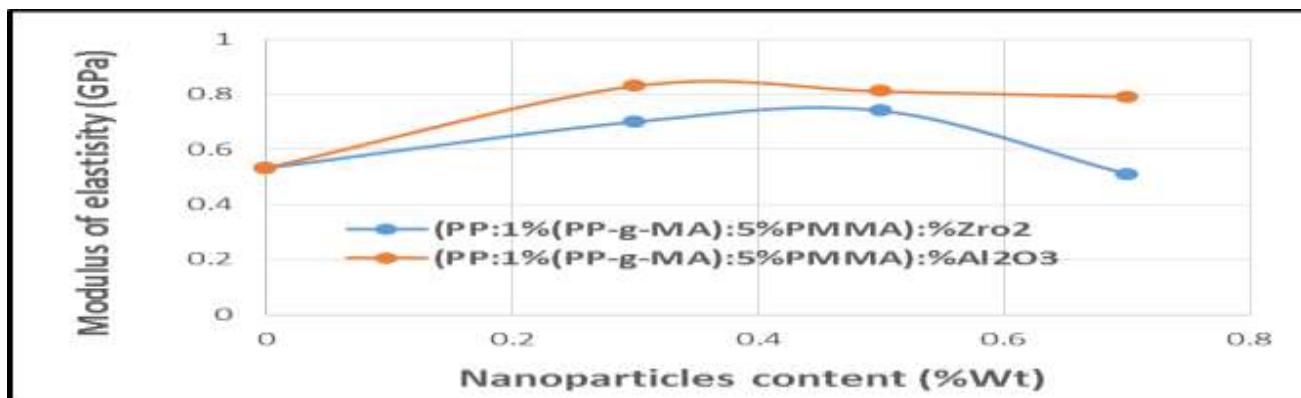


Figure 8: Elasticity modulus for Nanocomposites samples ((PP: 5%PMMA:1%(PP-g-MA)): X%ZrO<sub>2</sub> or X%Al<sub>2</sub>O<sub>3</sub>) as a function of Nanoparticle content in composite

The effect of adding both zirconia and alumina nanoparticles on the elongation at break for the nanocomposites is shown in Figure (9). It can be observed from this figure that the elongation at break decreased when adding zirconia and alumina nano powders.

This is due to the fact that zirconia and alumina are considered as rigid materials that led to reduce the ductility of the nanocomposites based on polymer blend (PP: 5%PMMA: 1%(PP-g-MA)), which has the high flexibility compared with the strong properties for zirconia and alumina [25].

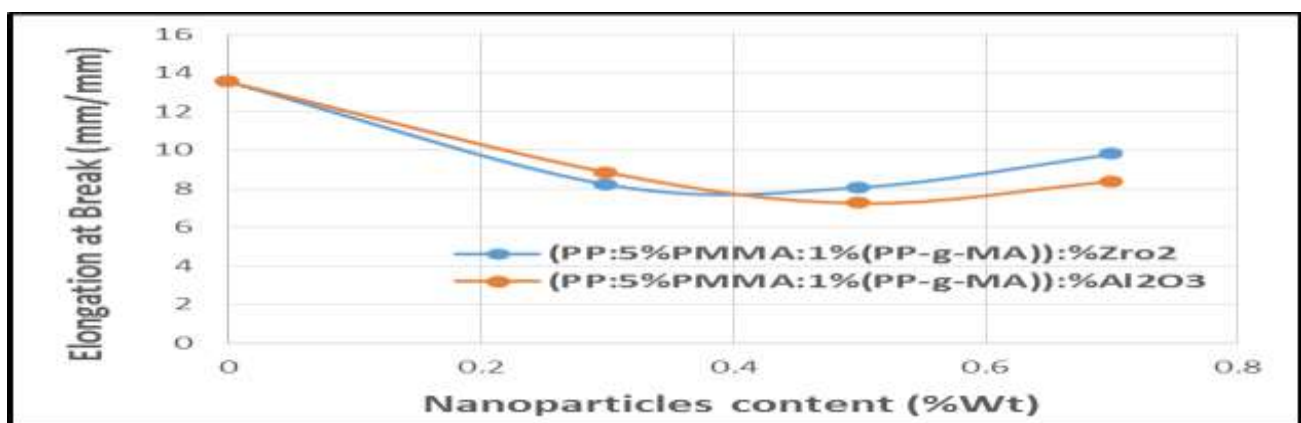


Figure 9: Elongation at break for Nanocomposites samples ((PP: 5%PMMA:1%(PP-g-MA)): X%ZrO<sub>2</sub> or X%Al<sub>2</sub>O<sub>3</sub>) as a function of Nanoparticle content in composite

### Flexural Test Results

The flexural strength and flexural modulus values of nanocomposites based on the polymer blend are manifested in Figures. (10) and (11), respectively. These figures indicated the slight increase in the values of flexural strength and flexural modulus with addition 0.3%wt. of (zirconia and alumina) nanoparticles to polymer blend (PP: 5%PMMA: 1% (PP-g-MA)).

At this ratio, was obtained the maximum values of flexural strength and flexural modulus were (81 and 81.5 MPa) and (1.93 and 1.95 GPa), respectively for the composite samples (PP: 5%PMMA: 1% (PP-g-MA)): % ZrO<sub>2</sub> or Al<sub>2</sub>O<sub>3</sub>). The increment of this ratio to larger than 0.3%wt. for both types of nanoparticle (zirconia and alumina) in composites ((PP: 5%PMMA: 1% (PP-g-MA)):

% ZrO<sub>2</sub> or Al<sub>2</sub>O<sub>3</sub>) led to reduce the values of flexural strength and flexural modulus for composites samples, this result is an agreement with previously reported in reference [26]. The negative effect in the flexural strength values when increasing the (zirconia and alumina) nanoparticles to larger than 0.3% wt. in composites may be regarded to the agglomeration of nanoparticles that occurred with high concentrations, especially in areas containing clusters of added nanoparticles with uneven distribution, these would act as centers of internal stresses concentration. So that, the resultant is a nanocomposite material with a low physical bonding between the nanoparticles and the components of polymer blend, and therefore, this requires low flexural stresses, to occur the failure, which leads to a lower flexural strength [24, 27].

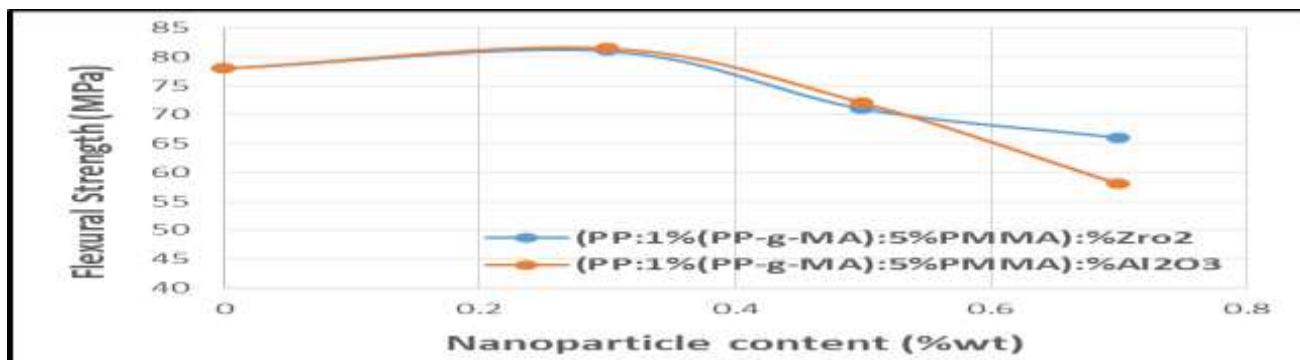


Figure 10: Flexural strength for Nanocomposites samples ((PP: 5%PMMA: 1 % ( PP-g-MA)): X%ZrO<sub>2</sub> or X%Al<sub>2</sub>O<sub>3</sub>) as a function of Nanoparticle content in composite

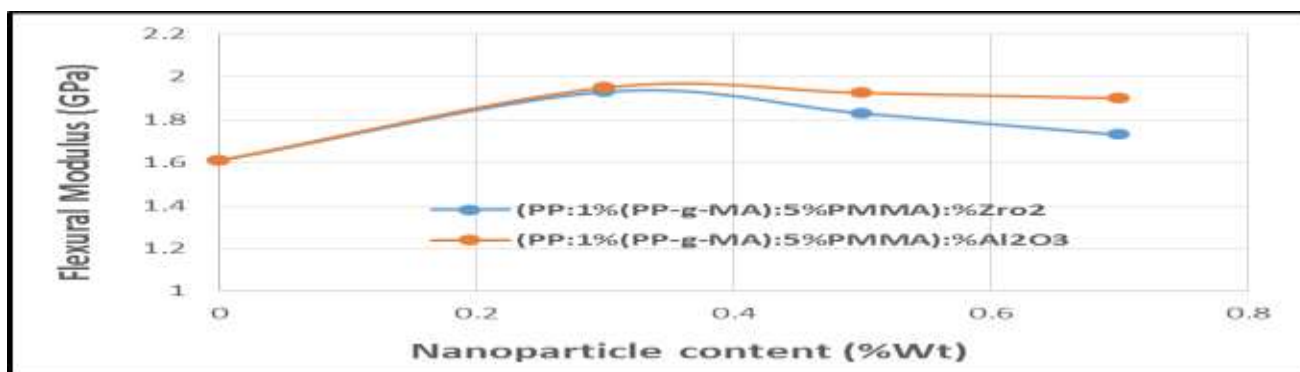


Figure 11: Flexural modulus for Nanocomposites samples ((PP:5%PMMA:1%(PP-g-MA)): X%ZrO<sub>2</sub> or X%Al<sub>2</sub>O<sub>3</sub>) as a function of Nanoparticle content in composite

## Morphology Test Results

Morphological information of the fracture surface for the neat polypropylene, polymeric blend and nanocomposites after a mechanically induced fracture from tensile test was studied by analyzing the photographs of scanning electron microscopy (SEM) at same magnification (3000X).

Figures (12 a and b) and (13 a, b, c, d) evince the various microstructures of morphologies of the fracture surface for the polypropylene, triple polymer mixture and its composites of (PP: 5%PMMA: 1%(PP-g-MA)): X% Zirconia or Alumina) respectively, depending on the polymeric blend components as well as on the type and ratio of (zirconia and alumina) nanoparticles content in these composites.

The SEM images of polymer blend (PP/PMMA/ (PP-g-MA) and its nanocomposites exhibited immiscible polymer blends. The morphology of immiscible polymer blends depends on the nature of components, ratios, processing conditions, wettability between their components and component melt viscosities [28]. In most heterogeneous systems, a morphology whereby one phase is distributed in another phase is observed. So, when the components of polymer blend composite are interchangeable (phase inversion), inversion

is important since at a certain ratio, the properties of composite, such as stiffness and impact strength, it may be change in an advantageous manner [29]. Where, the fracture surface of polymer blend (PP: 5%PMMA: 1%(PP-g-MA)) shown in Figure 12 (b) exhibited a homogeneous morphology, and this blend appeared to have a co-continuous morphology as strips and as a result, it was difficult to identify the individual polymers in this blend and composites.

The SEM photos of the fracture surface for the two groups of polymer blend composites aforementioned are shown in Figures 13 (a, b, c and d), respectively. It clearly shows a two-structure morphology, depending on the type and ratio of (zirconia and alumina) nanoparticles content in these composites. One of structures morphology was identified as a co-continuous structure, dense and homogeneous structural morphology nearly free from microscopic pores when added 0.3%wt. of (zirconia and alumina) nanoparticles in composite Figure 13 (a and b) displays smooth fracture surfaces, and this indicates to the better interfacial adhesion between polymer blend components and nanoparticle in composites which gave good mechanical properties for this type of the prepared composite samples, these surface



morphologies are similar to those appeared in [27, 30].

Whereas, the second structure morphology exhibited as strips, when alumina nanoparticles increased to 0.7%wt. in the composites, as shown in Figure 13 (d). Moreover, increasing the zirconia

nanoparticles to 0.7%wt. in composites (Figure 13 c)) led to increase the microscopic pores and micro-crack, which are indicated by red arrows, and this is may be causing the deterioration of the mechanical properties of the prepared composite samples when increasing the (zirconia and alumina) nanoparticles to larger than 0.5% wt. in composites.

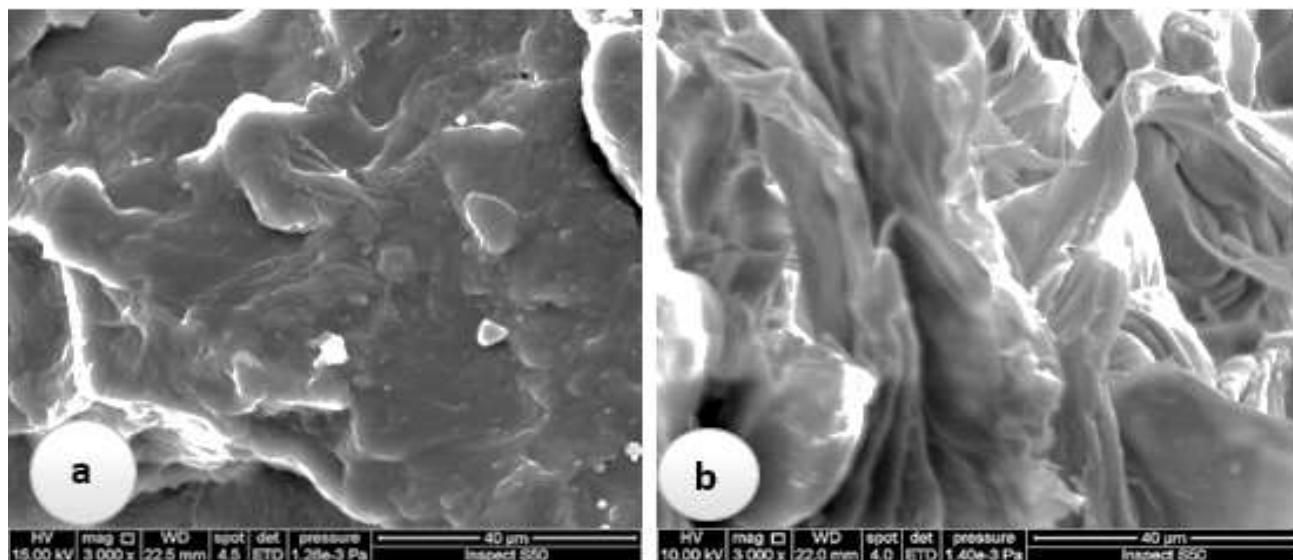


Figure 12: SEM image of fractured surface region of (a): Neat polypropylene and (b) ternary polymer blend (PP: 1% (PP-G-MA): 5% PMMA) at magnification (3000X)

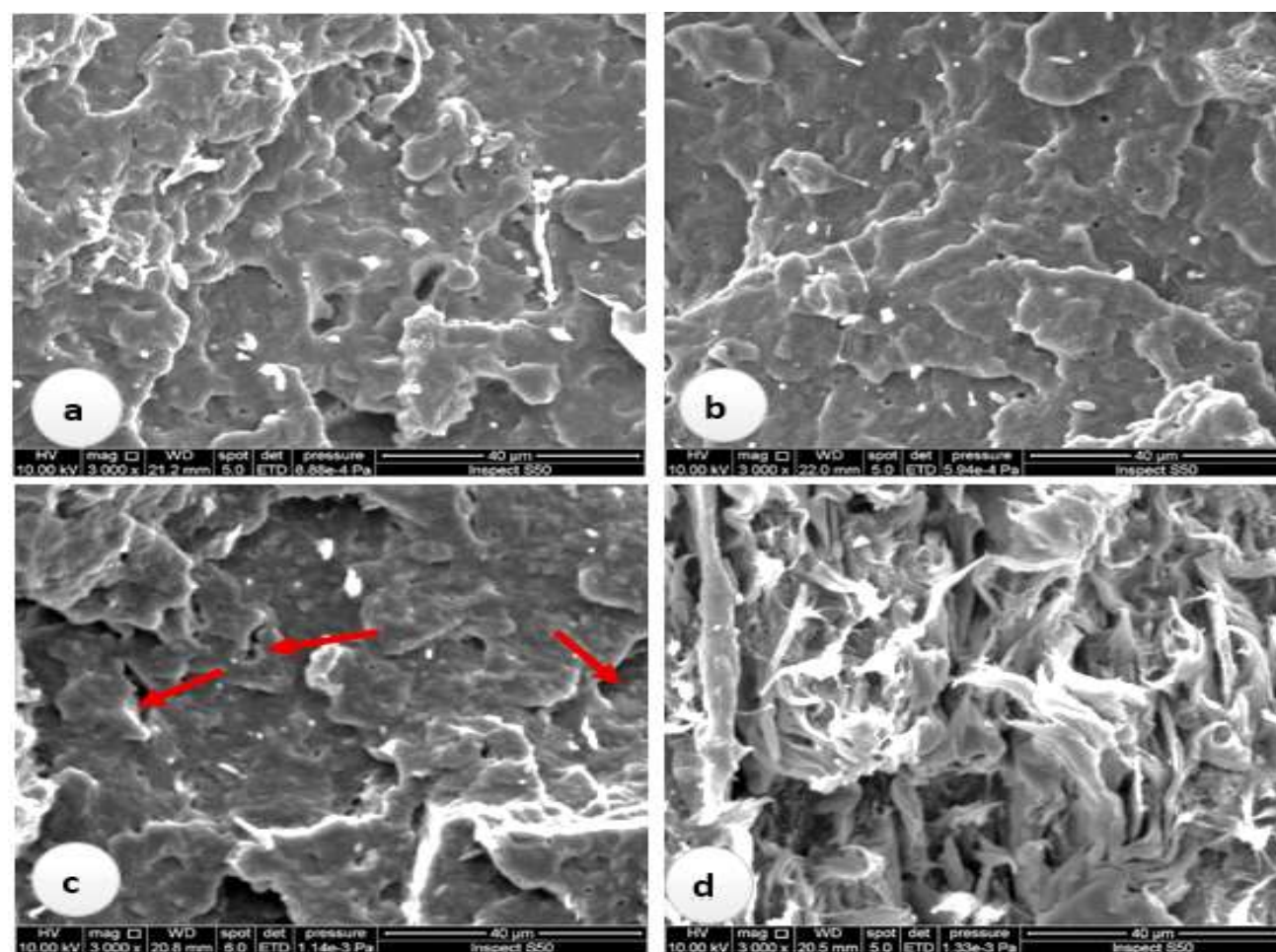


Figure 13: SEM photographs of the fracture surface morphology of nanocomposite as a function of nanoparticles content in composite (PP: 1% (PP-G-MA): 5% PMMA): X% ( $ZrO_2$  or  $Al_2O_3$ ). Where: (a): 0.3%  $ZrO_2$ , (b): 0.7%  $ZrO_2$ , (c): 0.3%  $Al_2O_3$  and (d): 0.7%  $Al_2O_3$ , at the same magnification (3000X)

## Conclusions

- FTIR test results pointed out to the prominence of physical bonding and absence of any chemical reaction between the components of the polymer blends and the nanocomposites as compared to the neat PP.
- Tensile properties and flexural properties of the polymer blend nanocomposite increased with the addition of any type of the nanoparticles (zirconia and alumina), except that the elongation at break decreased with nanoparticles.

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