Journal of Engineering Science and Technology
Vol. 3, No. 1 (2008) 97 - 108

THE INFLUENCES OF MELT-COMPOUNDING PARAMETERS ON THE TENSILE PROPERTIES OF LOW FILLER LOADING OF UNTREATED-MWCNTs-POLYPROPYLENE (PP) NANOCOMPOSITES

M.A.MOHD SALLEH [1], J. ABD. RAZAK [2*], N.A. IBRAHIM [3], A. FAKHRU'L RAZI[1], A.R. SURAYA[1]

[1]Chemical Engineering Department, Faculty of Engineering
[2]Chemistry Department, Faculty of Science
Universiti Putra Malaysia, 43400 UPM, Serdang, Selangor Darul Ehsan, MALAYSIA.
[3]Materials Engineering Department, Manufacturing Engineering Faculty, Universiti Teknikal Malaysia Melaka, Industrial Campus, 75450, Ayer Keroh, Melaka, MALAYSIA.
*Corresponding Author: jeefferie@utem.edu.my

Abstract

This study is to investigate the effects of addition self synthesised multi-walled carbon nanotubes (MWCNTs), to the final properties of polypropylene (PP) matrix nanocomposites. The influences of melt blending parameters were evaluated, where the interrelationship between the temperatures of compounding and roller rotor speed of shearing blade parameter, to the tensile properties of fabricated composites were studied. MWCNT was synthesised in the laboratory scale; by using the floating catalyst chemical vapour deposition (FC-CVD) method. Pre-compounding work is begun with de-agglomeration of MWCNT which carried out by combining the ultrasonication and mechanical stirrer means simultaneously. Carbon nanotubes produced was first verified by using SEM and TEM imaging microscopy techniques. It was later integrated with the thermoplastic PP matrix, via melt blending process through internally mixing approach. Very low weight percentage of chemically untreated MWCNT (0, 0.25, 0.50, 0.75 & 1.00 wt. %) was added into PP and later was compression moulded to the thin sheet of composites film. Composites were prepared by varying the compounding temperature into three processing temperature namely 165, 175 & 185°C and also into three shearing speed of roller rotor blade, 40, 60 & 80 rpm respectively. Later, it was mechanically tested via tensile testing following the ASTM D-638 standard method. The interrelationship between each parameter of compounding to the mechanical tensile properties was tested. It was shown that, the additional of very low loading of untreated-MWCNT filler content, does give moderate effects on reinforcement to the tensile properties of composite. Different compounding parameter gives significant difference to the pattern of plot which was comparable between each other.

Keywords: MWCNT, Polypropylene, FC-CVD, Melt Blending, Low Filler Loading, Untreated, Compounding Parameters.

1. Introduction

Polymer/CNTs composites have attracted considerable attention owing to their unique mechanical, surface and multi functional properties and strong interactions with the matrix, which resulting from the nano-scale microstructure and extremely large interfacial area [1]. Incorporation of CNTs into a polymer matrix can potentially provide structural materials with dramatically increased modulus and strength [2]. However, the reinforcing potential of CNTs is still widely undeveloped and needs further basic research. Assouline *et al*. cited from [1], found that the addition of 1 wt. % of MWCNTs into PP matrix increased the composites toughness due to the fibrillar crystal structure of PP induced by MWCNT [1]. Manchado *et al*. [3] found that CNT could be dispersed uniformly in PP by shear mixing, at minimum low filler loading, and they found that by adding 0.25–0.75 wt. % CNT into PP increased its tensile strength and stiffness as well as storage modulus considerably [3]. From this finding it can be claimed that, low amount addition of MWCNT into polymer matrix was still capable to improve its mechanical properties. Thus it will be tested in this study, by using the self synthesised MWCNTs which produced at the limited laboratory scale quantity. Polymer processing methods use large-scale quantities of raw materials, which are sometimes inappropriate when developing new composites based on costly filler such as CNT. Thus commercial MWCNT is not used due to the cost factor and this study was opt to use the low filler amount of loading (0 – 1 wt. %) of MWCNTs, since the study conducted by Breuer and Sundararaj [4] found that the low CNT loading of a few percentage was also significant in preserving the original physical properties of the polymer matrix [4].

To-date, the main challenges in CNT based polymer nanocomposites are to improve the uniform dispersion and alignment of CNTs within the polymer matrix, during the processing of these nanocomposites [1, 5]. CNTs are easy to agglomerate, bundle together and entangle, leading to many defect sites in the composites, and limiting the efficiency of CNTs on polymer matrices [1]. As an effort to solve these problems, this study has been emphasised the method of pre-compounding, in a way to pre-disperse the filler used. MWCNTs were first sonicated and mechanically stirred simultaneously in the chloroform solvent, prior to their incorporation with polypropylene matrix. Hence, the efficiency of the internal mixer machine in dispersing different weight percentage of untreated MWCNT within the polymer matrix will also be studied, where the melt-blending compounding parameters of temperature and rotation of roller rotor shearing blade were varied.

Melt mixing method via internal mixer technique is particularly desirable because of its speed, availability and its simplicity, especially in the common plastic industry. Resource from Breuer and Sundararaj [4], said that this method are quite advantage for nanomaterials addition to thermoplastic, especially for CNT and vapour grown carbon fibre (VGCF), since it will maintained the filler high aspect ratio and reduce the fiber breakage [4]. This method is also free from solvent and contaminant. Xiao *et al*. [6] reported that melt blending method can minimise the formation of CNT filler aggregates by applying the appropriate shear, during the melt mixing process. It is anticipated that compounding by using this route much probably will give better compounding effect which will result better mechanical tensile properties. However, back to the basic, effective utilisation of CNTs in the composites system was depending strongly on the

ability to disperse the CNT's individually and uniformly throughout the matrix without destroying their integrity or reducing their aspect ratio [4].

2. Materials and Methods

2.1 Raw materials

Polypropylene is used as matrix in this research is a product from TITAN PETCHEM (M) Sdn. Bhd., with commercial name of TITANPRO SM950. This type of polypropylene is classified as impact copolymer and has MFI value of 60g/10min at 230°C. The chemical used in this study was benzene (C_6H_6) with 99.5% purity. Benzene was used as a hydrocarbon source during synthesis of CNTs through FC-CVD process. Benzene has atomic mass of 78.11 g/mol. Ferrocene ($FeC_{10}H_{10}$) with 98% purity was used as a source of the ultra fine iron catalysts particles which reacted with benzene during chemical vapour deposition synthesis process. Ferrocene has a molecular weight of 186.04 with the brand name of ACROS ORGANICS. Two types of gases (supplied by MOX, Malaysia) were used; pure hydrogen (H_2) with 99.99% of purity and pure argon (Ar) with 99% of purity.

2.2 Preparation of MWCNTs by FC-CVD method

The chemical vapour deposition (CVD) method was employed for the synthesis of CNT. Girun *et al.* [7] described that this method is cheap and requires a relatively low deposition temperature compared to other techniques used to synthesise CNT. The parameter used to synthesis the MWCNT was 50 minutes reaction period, 850°C reaction temperatures, 350 ml/min hydrogen flow rate and 200 mg amount of ferrocene catalyst used. Argon gas was injected into the CVD reactor to prevent the oxidation of catalytic metal while raising the setting temperature. In order to produce the ferrocene vapour, the boat containing ferrocene which located at the edge of the ceramic tube, inside the CVD reactor was heated up to the temperature of 120°C, right after the reaction temperature had reached. After all the setting temperature became stable, hydrogen gases were bubbled into the benzene Erlenmeyer flask to form a mixed vapour of hydrogen and benzene for 50 minutes reaction period. Later, the furnace temperature was cooled down and then carbon nanotubes were collected from the ceramic boat and the wall of the ceramic tube.

2.3 Ultrasonication and mechanical stirring for MWCNTs filler

MWCNTs obtained from FC-CVD synthesis method were further dispersed by using sonication approach, prior to their compounding with polymer matrix. Sonication was done so that the carbon nanotubes were not entangled into the lumps and form flakes. Sonication was carried out for about 60 minutes in the solvent of chloroform and simultaneously was mechanically stirred for about 30 minutes until the solvent evaporated out (See Fig. 1 for ultrasonication-mechanical stirrer apparatus set-up). The carbon nanotubes sponge in the beaker was then dried overnight at 80°C in the drying oven. Later carbon nanotubes

powder was crushed by using an agate mortar. At this stage, it was assumed that, MWCNT is in the separate form between each other.

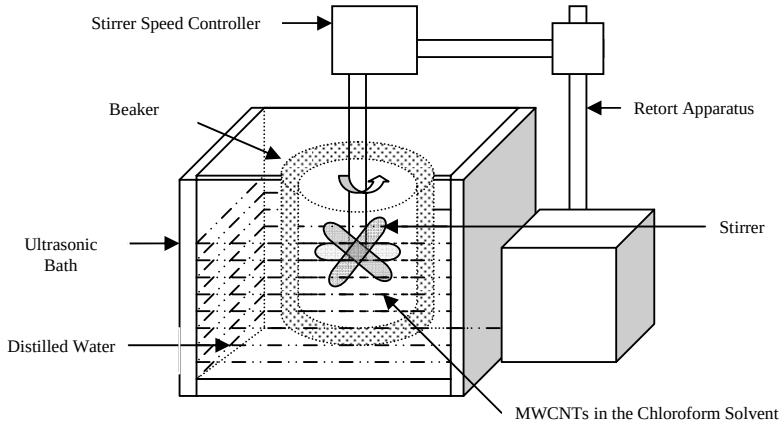

Fig.1. Ultrasonication-Mechanical Stirrer Apparatus Set-Up for MWCNTs Dispersion Procedure.

2.4 Characterisation of MWCNTs morphology by SEM & TEM

Morphology observation through SEM for the MWCNT, was performed on a Philips XL30 (Holland) Environmental Scanning Electron Microscope (ESEM). Samples of MWCNT were first adhered onto the surface of carbon tape and then mounted on the aluminium stubs which were then sputter-coated with a thin layer of gold by using sputter coater Polaron E-1500, in order to avoid the electrostatic charging phenomena during the observation. ESEM was operated at 25.0 KV accelerating voltage and signal was detected by secondary electron detector. Morphology observations through TEM for the MWCNT were performed on a Philips TEM-400 Transmission Electron Microscopy (TEM). Sample of MWCNT were first sonicated in alcohol solvent within 10 minutes, in order to disperse it. One drop of mixture was dropped properly onto the surface of *holey formvar* grid for negative staining and sample was dried prior their imaging.

2.5 Melt blending via internal mixer

Self synthesised MWCNT was pre-dried prior to weighing which carried out at 80°C for 24 hours in controlled vacuum oven. It was done purposely to expel any moisture from trapping within untreated MWCNT filler. All formulations were weighted based on the weight percentage from the total 40 gram per compounding. MWCNT-PP nanocomposites were prepared by melt blending in an internal mixer using the Thermo Haake PolyDrive - Rheomix R600/610 with various compounding parameters which summarised as in the following Table 1.

Table 1. Description of the Composites Compounding Parameters Designation Used.

Designation	Temperature	Rotor Speed
i	165	40
ii	165	60
iii	165	80
iv	175	40
v	175	60
vi	175	80
vii	185	40
viii	185	60
ix	185	80

In the preparation procedure, the thermoplastic PP were first loaded into the internal mixer and preheated for two minutes without rotation of roller rotor blade. Next eight minutes of compounding period were allocated and required amount of MWCNT (0, 0.25, 0.50, 0.75 & 1.00 wt %) were added to melted PP after four minute. The compounded recipes were then compression moulded using HSINCHU hot press size of 15x15 cm with the thickness of 1 mm for 10 minutes preheat and another 10 minutes compression period under a pressure of 150 kg/cm^2 at 185°C. The sheets obtained were immediately cooled between two plates of a water flow assisted cold pressed at room temperature for 10 minutes of cooling cycle.

2.6 Mechanical tensile testing

One millimetre thick dumb bell specimens were cut from the moulded sheets with a die cutter. Seven samples were tested for each composite formulation. Tensile tests were carried out according to ASTM D-638 on an Instron Universal Testing Machine 4302 with a load capacity of 1kN at cross head speed of 5 mm/min. The test was performed at 25±3°C in the humidity of 50%.

3. Results and Discussion

3.1 Morphological observation of synthesised MWCNTs

The microscopic study of the synthesised carbon nanotubes via FC-CVD method is shown as in Fig. 2. TEM observation with magnification up to 100 000 X (Fig. 2(a)) revealed that the carbon nanotubes produced by FC-CVD method are multi-walled type. The average internal and external diameters of hollow tube structure measured are between 10-25 nm and 30-55 nm respectively. With the thickness in that range for outer diameter, it can be claimed that the carbon nanotubes produced are multi-walled type. Length of MWCNT produced cannot be determined due to the nature of coiled and entangled structure of MWCNT. Variation of dimension for external and internal diameter of carbon nanotubes is due to the factor of yield collection after the synthesis process, where all the

collected CNTs were combined from all zones of ceramic tube and ceramic boat. Carbon nanotubes produced were not really straight in their structure and moderately long in their length. It is found that, MWCNTs produced were not really clean, wherein there is too many catalyst particle trapped inside the hollow tube, walls and at the edge of the tube. Observation through SEM with magnification of 10 000 X (Fig. 2(b)) shows that the carbon nanotubes produced were in the bundles, long structure and not well aligned. Presence of another carbonaceous product like amorphous carbon was detected through this observation. By increasing the magnification of SEM to 40 000 X (Fig. 2(c)), it can be clearly seen that the MWCNTs produced were open-ended type.

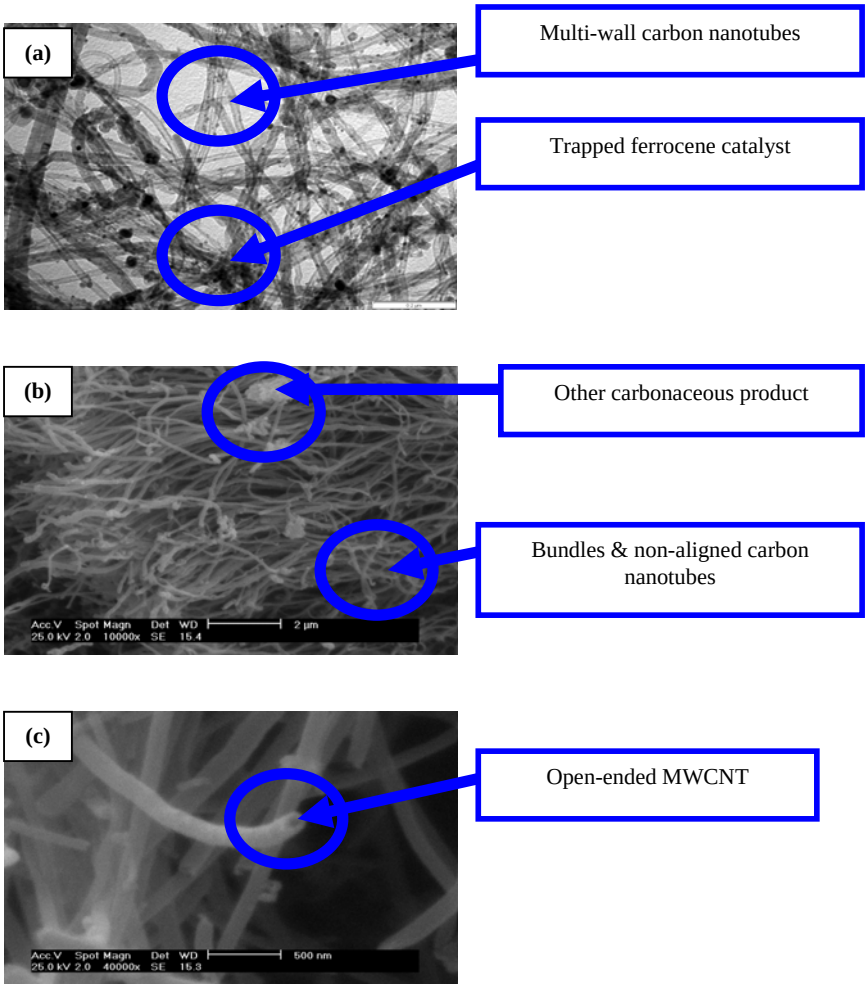

Fig.2. Morphology Observation of Synthesised MWCNTs through TEM & SEM.

3.2 Tensile properties evaluation

3.2.1 Tensile strength curves

Based on the bar-chart plotted (Fig. 3), there are three separations of categories in the graph. Effect of three compounding temperature in correlation with different rotation speed of roller rotor shearing blade at different compounding recipes (varied weight percentage of MWCNT) were studied. Group (i-iii) represent 165°C of compounding temperature, whereas group (iv-vi) and (vii-ix), represent compounding temperature of 175 and 185°C, respectively. From the first phase of the plotted bar-chart, it was shown that the overall tensile strength properties were monotonically increased with the increasing rotation speed of roller rotor blade over the amount of filler loading. Provided that, group i and ii shows the increment in the tensile strength properties up to the certain amount of filler loading, which are 0.75 and 0.50wt. % respectively, prior to their decrement in their subsequent amount of filler loading. Based from Gojny *et al*. [8], it is believed that the slight reduction in the tensile strength value at higher nanofiller contents can be attributed to an increasing amount of improper impregnated agglomerates, acting as imperfections in the composite, inducing early failure [8].

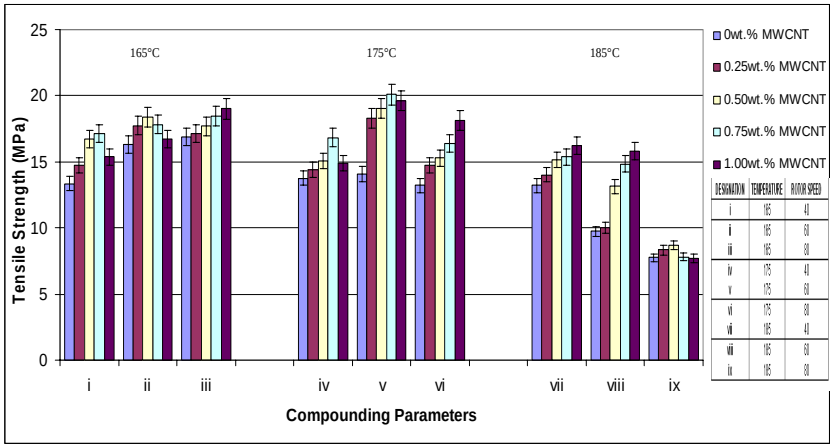

Fig.3. Tensile Strength of PP-MWCNTs Composites Compounded at 165-185°C at Various Roller Rotor Speed of Internal Mixer.

From the graph above we can see that, differences of increment value from the sample without addition of the MWCNT was significantly occurred at the middle part of the graph, where compounding temperature of 175°C were applied. Improvement in tensile strength properties were occurred for the composites from group (v). Distinct improvement happened, wherein addition of 0.25wt. % of MWCNT was increased the tensile strength value up to the 30% and addition of 0.75wt. % MWCNT contributed to the extreme enhancement until 42.8% of increment from 14.06 to 20.08 MPa. At the same group of compounding, addition of 1.00wt. % of MWCNT was decreased its tensile strength properties but the value plotted is still among the highest and significantly stronger than other fabricated composites, which is 19.62 MPa. This may be contributed by the factor

of nanotubes agglomeration which commonly occurred when it exist in large quantity in the compounding recipes.

Overall the pattern of the plot for the group (iv-vi) showed very good improvement in their tensile strength properties in comparisons with the un-filled matrix of PP polymer, where the most significant enhancement of tensile strength was obtained from the group of composites fabricated from CNTs which compounded at 175°C and 60 rpm which report an increment. The present results for the composites from group (vi) shows an almost linear increase of the measured tensile strength value in relation to the filler content, yielding at the maximum of 18.15 MPa at 1.00 wt. % MWCNT addition, under the given processing condition. This finding is in accordance with the previous work conducted by Park *et al*. [9], where in their study they found that, as CNT content increased, the tensile strength is increased gradually by the general rule of mixture. They claimed that polymer matrix added with CNT can increase the tensile properties and fracture energy because the crack propagation can be blunted by bridging up the crack faces [9]. Kueseng *et al*. [10] found that by incorporation of CNT even at low as 1.5% filler content, the tensile strength of nanocomposites was enhanced [10].

Application of compounding parameters for the group (viii) and (ix) was diminished its tensile strength properties of pure PP matrix, where the values plotted are lower than 10 MPa. Thus, it can be claimed that the temperature of 185°C with rotor speed of 60 and 80 rpm respectively is not suitable for MWCNT-PP composites fabrication. High temperature of compounding may expose the molecular chain of PP with the processing degradation, which generally will decrease its mechanical properties. With the exposure of high shearing rotation of roller rotor blade (80 rpm at 185°C), the effect of high temperature to the tensile properties of the fabricated composites becomes worst. This phenomenon can be seen at the group (ix), where all the plotted values are among the lowest, where the pattern of plot for this group of composites seems is level with no great improvement in its properties.

Thus, it can be concluded that temperature of compounding and roller rotor speed of melt blending, plays an important roles in the tensile strength properties of composites which containing lower amount of MWCNT filler loading. Both parameters are mutually correlates between each other and it should be highlighted as a main consideration during the development and production of carbon nanotubes based polymer nanocomposites in the real bulk manufacturing.

3.2.2 Modulus of Young curves

The Modulus of Young for fabricated composites is depicted as in Fig. 4. Samples were prepared at three different temperatures of various roller rotor speed of internal mixer, in five different compounding recipes for each group labelled, which varied from 0-1.00wt. % of MWCNT addition from total weight of compounding. Overall, by referring to the control sample (matrix of pure PP without addition of MWCNT) from each groups of compounding, it can be seen that the value of Modulus of Young were varied from 113.55 to 200.83 MPa, where the obvious trend of plot shown that, by increasing the rotation speed of the shearing blade, the pattern of the bar chart plotted were proportionally increased,

for groups (i-iii), monotonically increased for groups (iv-vi) and drastically decreased for the groups (vii-ix). Generally, for each group of compounding, addition of low amount of MWCNT was increased their value of modulus in comparison with the control sample, but only up to the certain level of amount before its properties dropped.

However, this is different with the group (vii), where the tested value of modulus was dropped when the 0.25wt. % MWCNT is added and increased again until the addition of 0.75wt. % MWCNT, before it dropped again at the addition of 1.00wt. % MWCNTs. It may due to the failure of compounding parameters (lower speed of roller rotor; 40 rpm); to fully disperse the MWCNT added which may create the formation of MWCNT agglomerates that tend to be the origin of flaw which may create failure, during the testing. The increment of modulus of young properties was not significantly occurred at the group (i-iii), where the temperature used is 165°C. The effect of roller rotor speed in this phase was not too obvious where the plot of the graph seems to be level. Great improvement of this property was occurred for all group of compounding in the second phase of the bar chart plotted (groups iv-vi) and only at the group (vii) and (viii) for the third phase of the graph. Highest increment of this property was occurred at the group (viii), where addition of only 0.75wt. % MWCNT, contributed to the enhancement of Modulus of Young value up to the 162.3%.

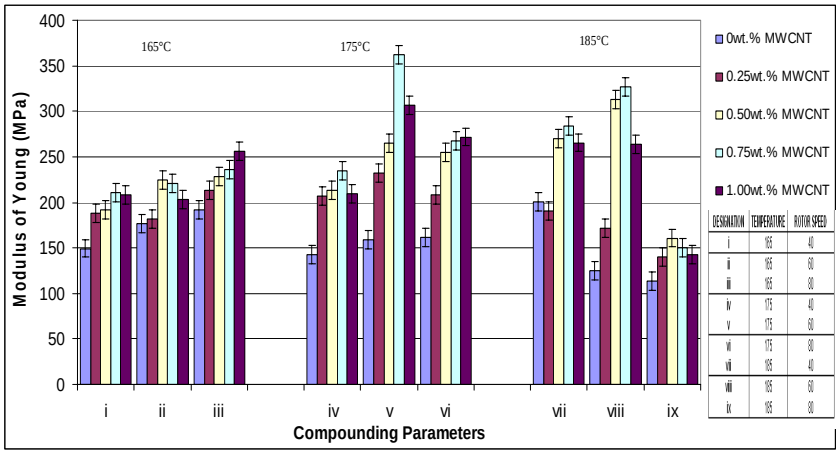

Fig.4. Modulus of Young of PP-MWCNTs Composites Compounded at 165-185°C at Various Roller Rotor Speed of Internal Mixer.

Kueseng *et al*. [10] found that the dispersion of nanoparticles in the polymer matrix used, gives rise to the increase in modulus. The increased stiffness may be caused by the formation of the immobilised matrix phases due to the incorporation of the nanotubes or nanoparticles [10]. The increment of modulus values with increasing the amount of filler loading was in agreement with what was reported by Girun *et al*. [7], where in high CNTs loading the nanocomposites are able to withstand more loads. The fabricated composites become stiffer. Maximum value plotted for this property occurred when the compounding using 175°C of compounding temperature at 60 rpm of rotor speed with the addition of

0.75 wt. % MWCNT was applied, where it improved the modulus of young value up to 128% in comparison with the pure PP which compounded at the same parameter of compounding.

Generally, using temperature of 185°C with high roller rotor speed of 80 rpm gives the value of modulus among the lowest compared to others. Again, it is revealed that this compounding parameter (group (ix)), is totally not suitable for the MWCNTs-PP nanocomposites fabrication. However, using this temperature but lowering the speed of shearing blade, still capable to give promising result of improvement in its properties for Modulus of Young.

3.2.3 Percentages of elongation at break curves

In overall, by increasing the temperature of compounding for each group, the percentages of elongation at break were decreased over the increasing amount of MWCNT filler loading. It can be seen that the pattern of plots for the first three groups in the first phase of the bar chart was proportionally decreased with the increasing roller rotor speed and the amount of filler addition. The decrement of elongation at break value in comparison with the value plotted for pure PP in each group from the first set (using 165°C), was not so difference.

Generally addition of lower filler amount caused the fabricated composites improved in their strength but diminished its capability to retain its ductility properties. The decreasing pattern of plot (Fig. 5) shows that the fabricated composites become brittle with the addition of MWCNT, even at the lower amount of addition. This finding is in agreement with the result obtained by Girun *et al*. (2007). They reported that the decrease in elongation at break with the increasing amount of CNTs that is rigid arises from the fact that the actual elongation experience by the polymer matrix is much greater than the measured elongation of the nanocomposites specimen [7]. Although the specimen is part of filler that is CNTs and part of matrix that is polymer PP, all the elongation is comes from the polymer if the filler is rigid. As the amount of CNTs is increased, the amount of polymer should be decreased. Thus, the decrement of elongation at break indicates that the incorporation of CNTs into polymer can improve the stiffness of the fabricated composites [7].

It can be seen that the addition of only 0.25 wt.% of MWCNTs drastically dropped the percentage of elongation at break until 33.6% of decrement. It happened to the composites which fabricated at the compounding temperature of 175°C using 40 rpm of rotor speed. In comparison with the pure PP compounded at the same parameter, PP/MWCNTs composites from the group (v) showed the decrement of the measured value in the range about 15.2 to 86.6%, which the pattern of decrement is among the stable without drastic pattern of sudden drop and quite distinct in their plot. Provided that, plot in the phase two of the bar chart for the group (iv) and (vi) showed that the plateau of measured value of elongation at break occurred at 0.5-0.75 wt.% and 0.25-0.75 wt.% of MWCNTs filler addition, respectively. The formation of plateau at both groups of composites shows that the use of high shearing speed of roller rotor blade at 80 rpm is not suitable for the fabrication of composites PP filled with low loading of untreated-MWCNTs.

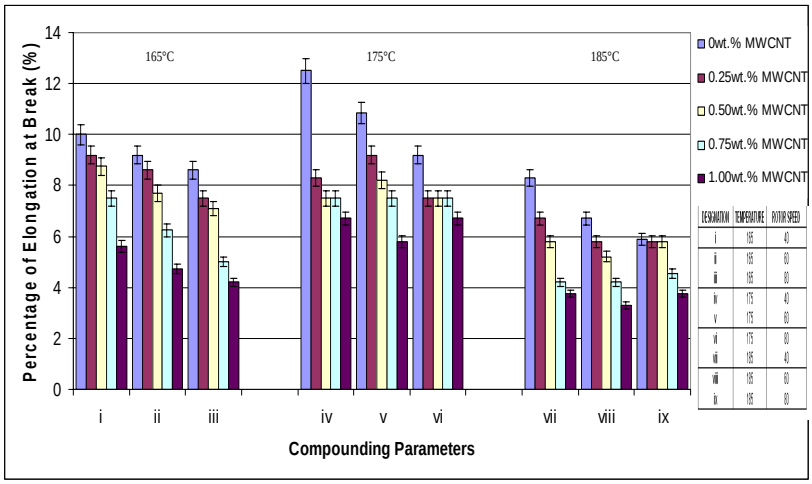

Fig.5. Percentages of Elongation at Break of PP-MWCNTs Composites Compounded at 165-185°C at Various Roller Rotor Speed of Internal Mixer.

The effect of compounding temperature at 185°C to the percentages of elongation at break for the PP/MWCNTs composites is not quite obvious for the groups of composites which compounded at 80 rpm of roller rotor speed of shearing blade. Varying the rotor speed value at the same temperature of compounding, still gives major influences to the strain properties, where it can be assumed that the discontinuance of PP polymer chain was occurred due to the high shearing effect of rotor blade. Generally, addition of MWCNTs will create deflation to the measured value of the percentage of the elongation at break due to the defect which may existed because of inhomogeneous dispersion of MWCNTs. The reduction in fracture strain was explained in terms of the existence of agglomerates, leading to local defects enhancing early failure [8].

4. Conclusions

In conclusion, parameter of melt blending like roller rotor speed of shearing blade and temperature of compounding, gives major impact to the tensile properties of low loading of untreated-MWCNT-PP nanocomposites. By increasing the rotor speed and temperature of compounding, the tensile strength and modulus of young properties initially will experiences the increasing pattern up to the certain level of filler addition before the properties diminish. However the percentages of elongation at break properties shows decreasing pattern of plots with the increasing amount of MWCNTs filler addition. In overall, it can be concluded that the composites fabricated by using the compounding temperature of 175°C and rotor speed of 60 rpm which comes from group (v) composites, provides the best performance of experimental tensile properties, in comparisons with other groups of composites which fabricated by using other compounding parameters.

References

1. Xie, X.L., Mai, Y-W., and Zhou, X-P. (2005). Dispersion and alignment of carbon nanotubes in polymer matrix: A review. *Materials Science and Engineering R,* 49, 89-112.
2. Ming Li Z., Ni Li S., Bo Yang M. and Huang R. (2005). A novel approach to preparing carbon nanotubes reinforced thermoplastic polymer composites. Letters to the Editor / *Carbon*, 43, 2397-2429.
3. Manchado, M.A. Lo'pez, Valentini, L., Biagiotti, J., and Kenny, J.M. (2005). Thermal and mechanical properties of single-walled carbon nanotubes-polypropylene composites prepared by melt processing. *Carbon*, 43, 1499-1505.
4. Breuer, O., and Sundararaj, U. (2004). Big returns from small fibers: A review of polymer / carbon nanotubes composites. *Polymer Composites*, 25(6).
5. Sandler, J., Werner, P., Shaffer, M. S.P., Demchuk, V., Altstadt, V., and Windle, A.H. (2002). Carbon-nanofibre-reinforced poly (ether ether ketone) composites. *Composites: Part A,* 33, 1033-1039.
6. Xiao, K.Q., Zhang, L.C., and Zarudi, I. (2007). Mechanical and rheological properties of carbon nanotubes-reinforced polyethylene composites. *Composites Science and Technology* 67:177-182.
7. Girun, N., Fakhru'l-Razi A., Suraya, A.R., and Muataz, A.A. (2007). Multi-wall carbon nanotubes/styrene butadiene rubber (SBR) nanocomposites. *Fullerenes, Nanotubes and Nanostructures*, 15(3), 207-214.
8. Gojny, F.H., Wichmann, M.H.G., Fiedler, B. and Schulte, K. (2005). Influence of different carbon nanotubes on the mechanical properties of epoxy matrix composites – A comparative study. *Composites Science and Technology*, 65, 2300-2313.
9. Park, J.-M., Kim, D.-S., Lee, J.-R., and Kim, T.-W. (2003). Nondestructrive damage sensitivity and reinforcing effect of carbon nanotubes/epoxy composites using electro-micromechanical technique. *Materials Science and Engineering C,* 23, 971-975.
10. Kueseng, K., & Jacob, K.I. (2006). Natural rubber nanocomposites with SiC nanoparticles and carbon nanotubes. *European Polymer Journal,* 42, 220-227.