

PAPER • OPEN ACCESS

Effect of filler size and loading on thermo-mechanical degradation of polypropylene-ethylene/wollastonite composite

To cite this article: Lip Teng Saw *et al* 2020 *IOP Conf. Ser.: Mater. Sci. Eng.* **864** 012114

View the [article online](#) for updates and enhancements.

You may also like

- [Zirconia-containing wollastonite ceramics derived from biowaste resources for bone tissue engineering](#)
Palakurthy Srinath, P Abdul Azeem, K Venugopal Reddy *et al.*
- [Sputtering Behavior of Rough, Polycrystalline Mercury Analogs](#)
H. Biber, J. Brötzner, N. Jäggi *et al.*
- [Wollastonite-based asphalt concrete](#)
T I Shishelova, V V Fedchishin and M A Khramovskih



The Electrochemical Society
Advancing solid state & electrochemical science & technology



249th
ECS Meeting
May 24-28, 2026
Seattle, WA, US
Washington State
Convention Center

Spotlight Your Science

**Submission deadline:
December 5, 2025**

SUBMIT YOUR ABSTRACT

Effect of filler size and loading on thermo-mechanical degradation of polypropylene-ethylene/wollastonite composite

Lip Teng Saw¹, Firuz Zainuddin¹, Cao Xuan Viet² and Du Ngoc Uy Lan^{1,3}

¹School of Material Engineering, Kompleks Pusat Pengajian UniMAP, Taman Muhibbah, University Malaysia Perlis, 02600 Jejawi, Perlis, Malaysia.

²Faculty of Materials Technology, Ho Chi Minh City University of Technology, Vietnam

³Department of Polymer Engineering, Universität Bayreuth, Germany

E-mail: Du.Uy-Lan@uni-bayreuth.de,epoxylan@gmail.com

Abstract. This study is to understand the side effect of wollastonite fillers in polymer compounding that caused different levels of thermo-mechanical degradation (TMD) during extrusion. Wollastonite has a needle-like structure with different ratio of L/D; it compounded with the polypropylene-ethylene copolymer (PPcoE) using a twin-screw extruder. The effect on the TMD of PPcoE was determined by analysing the by-products within the extrudates via FTIR. The rheology of PPcoE composite was measured using a capillary rheometer, and the thermal stability of composite was studied using TGA. In this study, TMD is classified as two levels: chain scission and oxidisation. The wollastonite filler were found to accelerate the TMD mechanism and generated oxidised products during extrusion. The reduction of filler size effectively minimised the chain cleavage of PPcoE matrix, also come with improvement in thermal stability. However, increasing the filler loading had a dominant effect of inducing more chain cleavage on the polymer matrix.

1. Introduction

Polypropylene-ethylene copolymer (abbreviated as PPcoE) is a common engineering thermoplastic with better manufacturability and elastic properties over homo-polypropylene. The addition of mineral fillers into PPcoE matrix provided higher stiffness and thermal resistance properties, better productivity and surface appearance, as increasing the commercial values of PPcoE product. The PPcoE composites usually compounded via extrusion and then manufactured using the injection-moulding for large-scale and time-efficient production purpose [1,2]. These processes require the PPcoE matrix undergo a molten state whereby the PPcoE blended with the solid particles, and then the mixture could be deformed or compressed into the desired shape. Thus, both heat and mechanical forces were applied to the PPcoE matrix during the melt-process; the heat source is to soften the polymer matrix, and the mechanical forces are to convey the polymer-filler mixture toward the chamber's exit, meanwhile also mingling them [1, 3]. However, the polymer chain tends to undergo mechanical cleavage during the melt-processing, which the polymer chain was stressed and broken by a tension force. The tension force could be originated from the polymer-wall shearing activity (when the polymer chains contacted and dragged over an equipment's wall/screw/injection ram surface), or from the entanglement and dragging activity of two polymer chains. This breakage of polymer chain generated reactive end-group, which may cause the polymer to undergo degradation and lowering the product's properties. The heat energy during melt-



processing only enough for melting purpose, which the processing temperature is usually lower than the polymer actual decomposition temperature, nevertheless, the heat source may accelerate the degradation of a polymer chain that was mechanically cleavage [4–6]. This type of degradation also claimed as thermo-mechanical degradation (abbreviated as TMD).

In the aspect of processing parameters, there are well known that both processing temperature and speeds (e.g. screw and injection speed) has a proportional effect on TMD; higher temperature/speed results in heavier degradation [7–9]. The additional processing aid or modify the processing environment give an advance to minimise the degradation of polymer during melt-processing. Such as the usage of antioxidant and the mixing chamber purged with nitrogen gas, both are claimed to be effectively lowering the oxidation of polymer to occur during melt processing [10, 11]. In the view of polymer composite compounding, the addition of filler particles into the polymer matrix will increase the viscosity of the melted polymer composite, which also increase the difficulty of polymer chains movement [12, 13]. Theoretically, this will induce heavier TMD since the solid particles will generate more stress concentration points within the melt polymer matrix and causes more rapid chain cleavage. However, it still lacks evidence and studies to support this proposition.

On the other hand, the chemistry of filler also gives an impact to the polymer stability during melt-processing. Many research reported the decomposition of organic filler and certain inorganic filler (e.g. zeolite) during melt-processing results in the formation of oxidised products within the polymer, also lower molecular weight [14–19]. But when come to mineral filler, this consideration is usually ignored by many researchers, since mineral fillers were majorly claimed to be thermally stable and inert [12, 14, 15]. Nevertheless, the hydroxyl surface of mineral filler supposedly has a role on the melted polymer stability; the hydroxyl surface could accelerate the oxidation of polymer during melt processing, which acts as a heterogeneous catalyst between oxygen molecules and cleaved polymer chains. The author has a similar find out from the previous study: the melt-compounding of kaolin filler (aluminium silicate) and polypropylene copolymer produced oxidised side products, e.g. alcohol, aldehyde, ketone and ester. These side products concentration proportionally increase along with the kaolin filler loading [16, 17]. Yet, more experimental work of other type mineral filler composite is required to verify this perspective. Therefore, a similar study was carried out in this paper by replacing the kaolin filler with wollastonite filler (calcium silicate) to investigate the catalyst effect of hydroxyl surface, and also to evaluate the influences of filler size and filler loading on the TMD of PPcoE composite.

2. Experimental

2.1. Material

The polypropylene copolymer supplied from Titan Polymer (M) Sdn.Bhd. with grade SM-340. It has a melt flow index value of 4 g/10mins under standard ASTM D1238. Wollastonite, CaSiO₃, obtained from Kaolin Sdn.Bhd., Malaysia. This filler has a feature of needle/rod shape and loss ignition of 4±1% at 1025°C. Two filler sizes were selected: KM25W grade has a longer and larger particle (lower than 60 µm) with a 13:1 length to diameter ratio; grade KM45W has a shorter length and smaller particle size (lower than 40µm) with a 4:1 length to diameter ratio. The SEM figure of these two wollastonite grades was shown in figure 1.

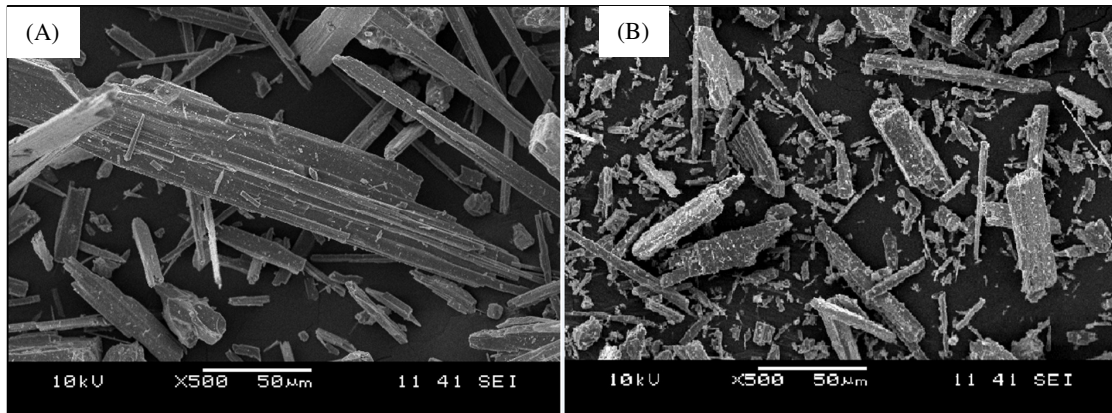


Figure 1. (A) KM25W and (B) KM45W

2.2. In-Situ Epoxidation

The PPcoE/Wollastonite composite was compounded using a twin-screw extruder, model Benchop 16-40 from Labtech Engineering Company. The twin-screws have an inter-mesh and co-rotational mode, 16mm diameter and a ratio of 40 L/D. All ingredients were dried at 80°C for 12 hours before the extrusion process. The processing temperature set at 190°C for all heating barrel, screw speed fixed at 50rpm. All ingredients were pre-mixed according to their formulation before feed into the extruder hopper, these formulations shown in table 1. The extrudates were pulled and go through a water bath at room temperature (25±2°C) for cooling purpose. Then, the solidified extrudates were continuously pulled into a pelletiser machine and cut become pellet form. These pellets were dried using a hot-air circulated oven, 80°C for 4 hours before any further testing.

Table 1:The formulation of PPcoE/Wollastonite composites.

Sample	PPcoE, wt%	KM25W, wt%	KM45W, wt%
Control	100	0	0
PPcoE/W10S	90	10	0
PPcoE/W20S	80	20	0
PPcoE/W10L	90	0	10
PPcoE/W20L	80	0	20

2.3. Testing and Analysis

2.3.1. Capillary Rheology Test

The capillary rheology test was carried using the Dynisco LCR-7001 rheometer at 230°C and a 2.5-1000s⁻¹ shear rate range, the operating standard was referred to the ASTM D3835. The change of pressure recorded, then the viscosity was calculated using the built-in software. These data were collected and replot into a log10 scale graph for a better observation purpose.

2.3.2. Thermogravimetric Analysis

The thermal-oxidative degradation test was carried on a TGA-Q500 instrument from 25°C to 600°C with 10°C/min heating rate, the operating standard was according to ASTM D2584. The decomposition temperature at 50wt% (T_{50%}), the end decomposition temperature (T_{end}), the onset decomposition temperature (T_D) and the decomposition rate (D_{slope}) were determined from the TGA curve. TD was

obtained from the intersection point of two tangents from the region of thermograms before and during decomposition. The temperature at the maximum decomposition rate, T_{Dmax} , was obtained from DTG.

2.3.3. Fourier Transfer Infrared Analysis

The FTIR machine used in this research branded Perkin Elmer Spectrum RX1 PC Ready LX185256 assembly with a Pike Miracle™ Single Reflection Horizontal ATR. The testing parameters were set to 4 cm^{-1} resolutions, 2 cm^{-1} intervals, and 16 times of scanning at the range of $4000\text{--}600\text{ cm}^{-1}$. All spectra were smoothed, and peak height was measured using Perkin Elmer's Spectrum software. The Beer-Lambert law was used to investigate the concentration of a polymer functional group; however, due to the commercial confidential, the actual concentration of the functional group is unknown. Therefore, the intensity ratio was used to roughly estimate and indicate the functional group amount by comparing two peaks intensity with unknown concentrations. The intensity ratio is calculated using the formula of $i_{\text{product}}/i_{1465}$, where i_{product} refers to the intensity or peak height of a degraded product, and i_{1465} refers to the peak height of the peak at 1468 cm^{-1} , which corresponds to the molecular motion of $-\text{CH}_2-$.

3. Results and Discussions

3.1. The rheology of PPcoE/Wollastonite composites

Figure 2 displays the viscosity of PPcoE/Wollastonite composites. The linear decrement trends of all formulation indicate the pseudo-plastic behaviour of thermoplastic that was shear-thinning took part along with the increment of shear rate, and the addition of wollastonite filler does not change this nature. However, the filler size and loading give an impact to the viscosity value of composite. PPcoE/W20L shows slightly higher viscosity than PPcoE/10L at low shear rate range below than 10 s^{-1} , but both composites have almost alike flow behaviour after 25 s^{-1} (the zoom-in portion). This finding fit to a common agreement that the increment of filler loading leads to a difficult melt flow, whereby those filler particles obstructing the melted polymer movement and require higher pressure to achieve the flow rate [12, 13]. Yet, due to the needle-like nature of wollastonite, the composite reached a critical point at 25 s^{-1} , where those particles have sufficient energy to align with the flow direction, allowing the melted polymer to carry it and flow together smoothly. This phenomenon also detected in other formulation, mainly at shear rate range after 500 s^{-1} , the difference in viscosity of each composite at this range is very closeby. PPcoE/W10S was found to have a very similar flow behaviour as control, and this could be due to the small needle particles have the advantage to be aligned. But when the filler loading increased to 20wt%, this advantage was surpassed by the obstruction effect of the massive immobile solid phase.

3.2. The thermal stability of PPcoE/Wollastonite composites

The thermal stability of PPcoE/Wollastonite composites was displayed in figure 3, and the detail of thermal properties was summarised in table 2. The TGA thermograms propose the thermal stability of control have higher than those wollastonite filled composites, and this could be attributed to the earlier decomposition of the oxidised or low molecular weight products within the PPcoE matrix [22, 23]. Moreover, the weak interphase between the polymer matrix and wollastonite filler also weaken the thermo-oxidative resistance of the polymer phase. The weak interphase does not provide any physical entrapment advantage to hold the polymer chain against heat and oxidation, then cause an earlier initiation of decomposition. However, due to its wellly dispersed wollastonite phase, those discontinuous phases have acted as a multi-layer of the heat separation barrier [25], and prolonged the decomposition of polymer matrix whereby results in lower D_{slope} value.

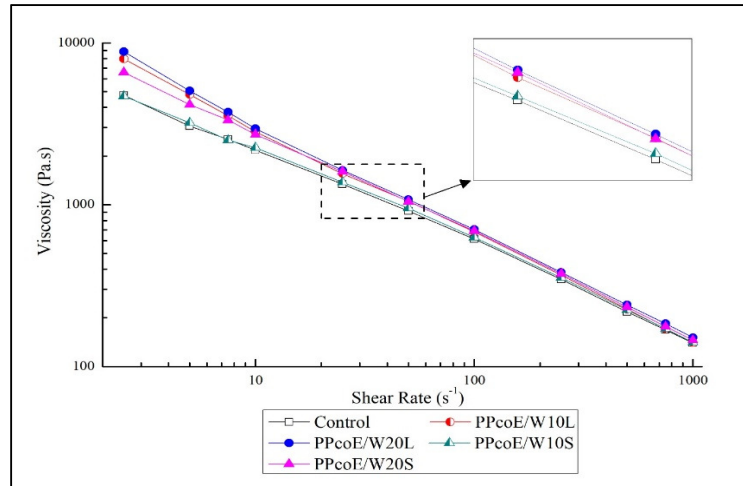


Figure 2. The rheology result of PPcoE/Wollastonite composites.

The T_{onset} of PPcoE/W20L is 103% higher than PPcoE/W10L, and the T_{onset} of PPcoE/W20S is 102% higher than PPcoE/W10S. This fits the general agreement that higher the filler loading, higher the thermal stability of the composite [25, 26]. But in the aspect of filler size, the smaller particle-filled composite has the better properties, and the increment is around $5.25 \pm 0.35\%$ when reduced in size. The smaller wollastonite particle not only advances in distribution and dispersion, but it could substantially preserve the polymer stability during the extrusion process. As mentioned in rheology section, wollastonite filler has the easy realign benefit during melt compounding, and smaller particles size would allow the melted composite to convey more effortlessly. The machine applied less torque and pressure to the PPcoE/W10S and PPcoE/W20S; thus both composites have higher thermal stability than PPcoE/W10L and PPcoE/W20L since the TMD of the composite was minimised.

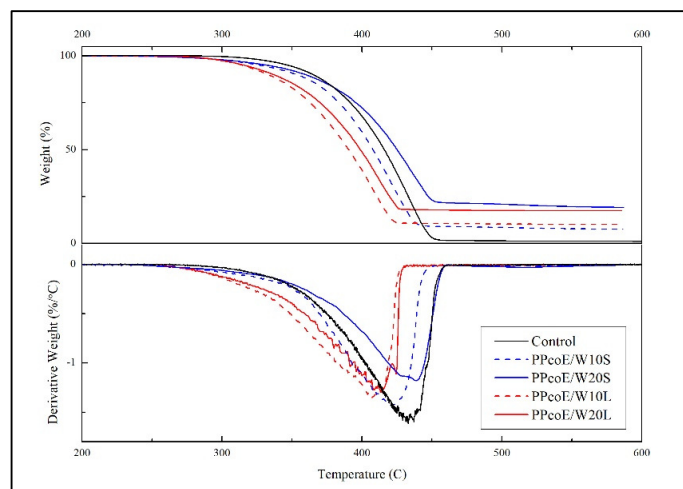


Figure 3. The TGA and DTG thermograms of control and PPcoE/Wollastonite composites

Table 2: The detailed TGA and DTG summary of PPcoE/Wollastonite composites.

	T_{onset} (°C)	$T_{50\%}$ (°C)	T_{end} (°C)	T_{Dmax} (°C)	D_{slope} (wt%/°C)
Control	378.0	416.0	461.5	432.4	-1.378
PPcoE/W10L	347.1	390.5	435.4	407.0	-1.083
PPcoE/W20L	358.6	399.0	435.3	415.6	-1.050
PPcoE/W10S	366.8	408.0	456.5	421.1	-1.267
PPcoE/W20S	376.5	425.5	462.0	437.0	-1.106

3.3. The degraded products

Figure 4 displayed the FTIR spectrum for each formulation, and detail of those plotted labels were summarised in table 3. There were clear that the control has no any oxidative product; the dominant side products were alkene end-group which from the disproportionation during TMD, the mechanism was proposed as figure 5. When focusing on filled composites, the oxidised products were detected. This result suggests that the presence of wollastonite filler was accelerated the TMD of polymer and lead to oxidative degradation, which also proved the hypothesis of this work – the hydroxyl surface of wollastonite act as a heterogeneous catalyst in TMD. There were ester and carboxylic groups were found within the filled composites, which suspected due to the further oxidation between alcohol and aldehyde side-products that generated during TMD (proposed as figure 6).

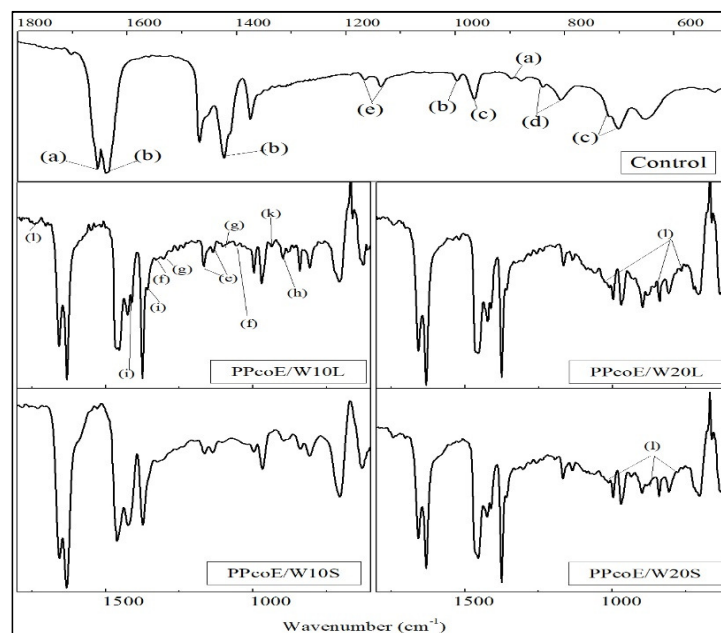


Figure 4. The FTIR spectra of control and PPcoE/Wollastonite composites.

Table 3: The functional groups of degraded products during TMD and their reference wavenumbers.

Type of Stabilization		Functional Group Name	Reference Wavenumber, cm^{-1}
Disproportionation	(a)	Alkene endgroup with 2,2 di-substituted	1655±5, 1415±5, 890±5
	(b)	Alkene endgroup with mono-substituted	1640±5, 1415±15, 990±5
	(c)	Alkene endgroup with di-substituted	trans 970±5, 695±35 Cis 700±30
	(d)	Alkene endgroup with tri-substituted	1670±5, 865±25 (vibration H from -CH= and R) 1675±5, 825±25 (vibration H from -CH=)
Hydrogenation	(e)	Di-methyl endgroups	1370±5, 1160±20, 1125±25
Oxidation	(f)	Primary Alcohol	1045±15, 1305±45
	(g)	Secondary Alcohol	1305±45, 1105±20
	(h)	Aldehyde	880±100
	(i)	Ketone with a Methyl endgroup	1365±15, 1425±25
	(j)	Carboxylic acid	1710±15, 925±25
	(k)	Ester	1740±10, 1240±10
	(l)	Tertiary Alcohol	745±25, 1175±25, 1020±20, 870±50

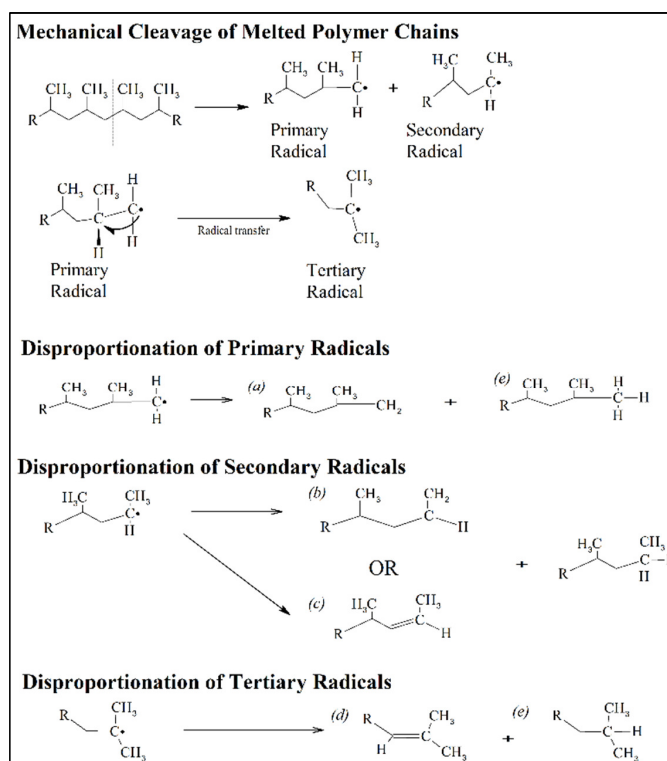


Figure 5. The mechanical cleavage of PPcoE and the disproportionation mechanism

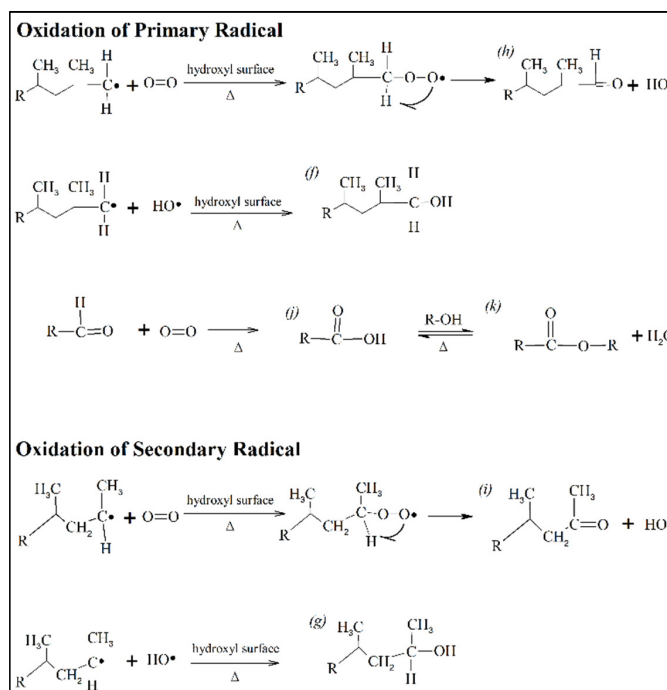


Figure 6. The oxidative stabilization of TMD

When the filler loading increased to 20wt%, the PPcoE/Wollastonite composites shown detection of a tertiary alcohol functional group, this result implied that the tertiary radical might tend to oxidative propagate to an alcohol species only when the reaction energy/condition is sufficient. The effect of filler size on TMD degraded product was illustrated as figure 7, and smaller particle has a dominant effect to minimise the filler drawback on TMD. The intensity ratio for overall degraded products of PPcoE/W10S has slightly lower than PPcoE/W10L, except the aldehyde group which the chemical residual of oxidation that was interrupted by the extrusion/cooling process. The extreme low in the peak intensity ratio of the carboxyl acid group has complied to this assumption.

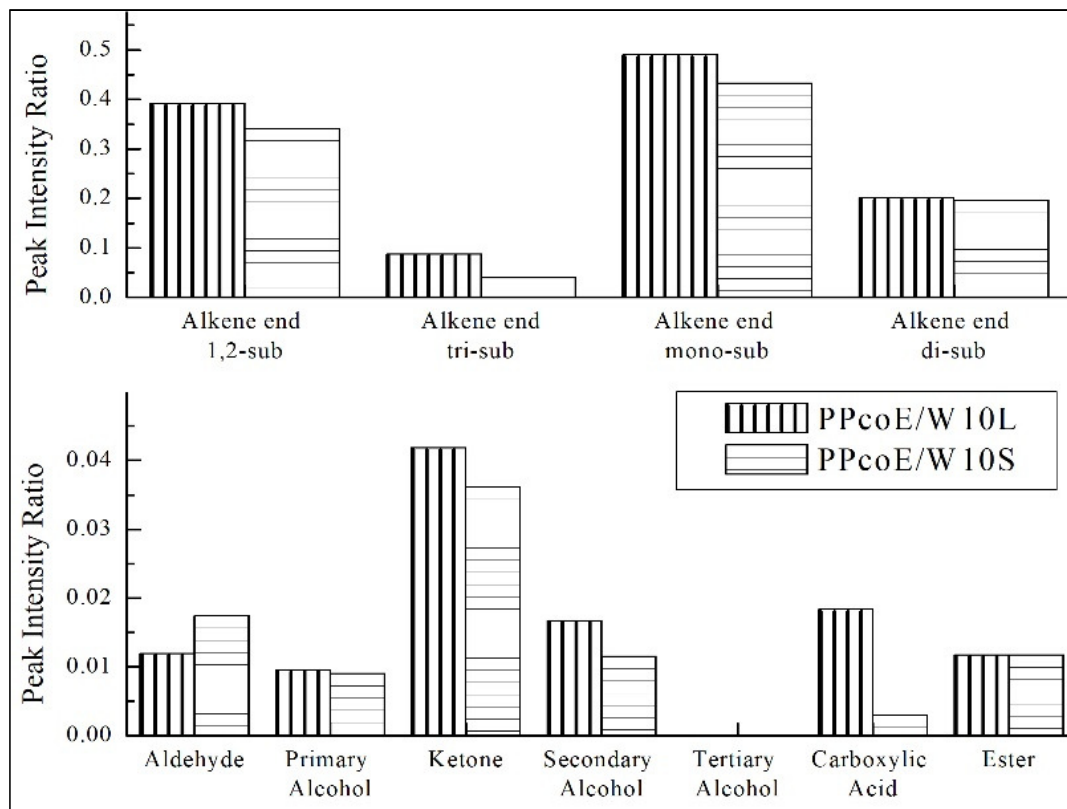


Figure 7. The comparison of peak intensity ratio for each functional group within PPcoE/W10L and PPcoE/W10S.

Figure 8 indicates the peak intensity ratio of oxidised products for PPcoE/Wollastonite composites when the filler loading increased from 10wt% to 20wt%, a significant increment in primary alcohol, ketone and ester group for both filler size filled composites. This also suggests the higher filler loaded composites were suffered from heavy oxidation during the extrusion process. The formation of tertiary alcohol also one of the evidence to support this hypothesis.

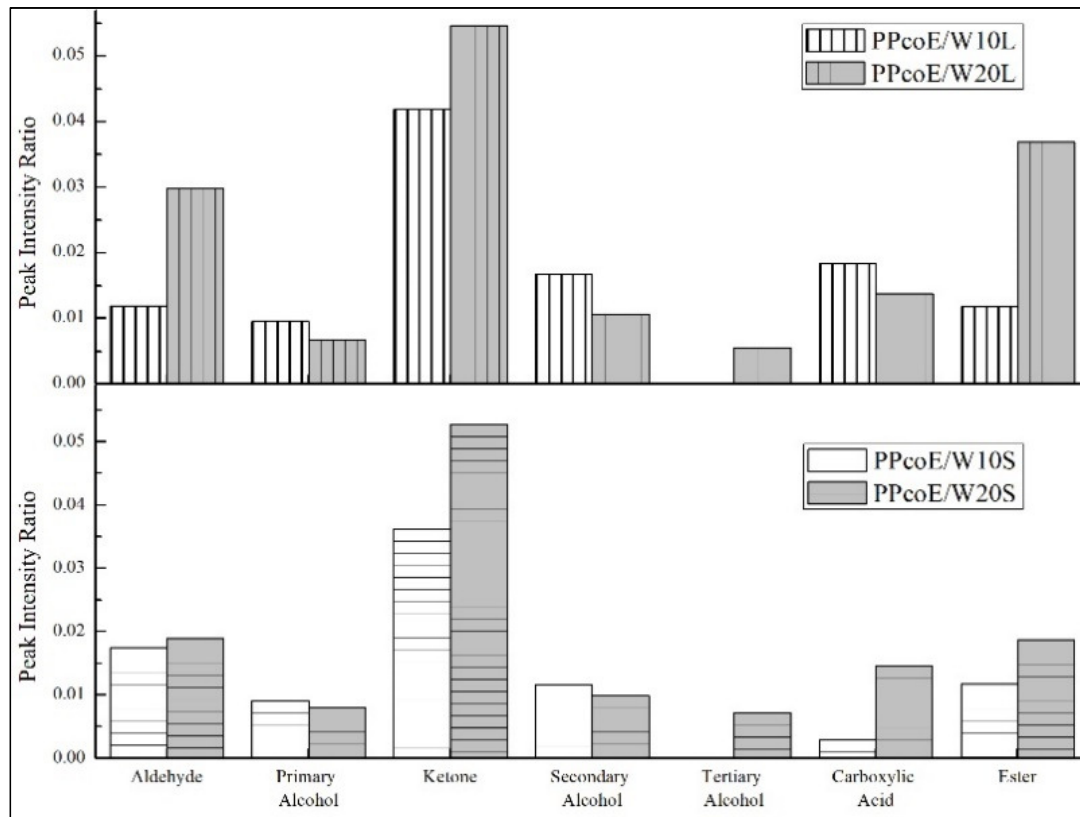


Figure 8. The comparison of peak intensity ratio of oxidised products within PPcoE/Wollastonite composites.

4. Conclusions

The realignment nature of needle-like wollastonite particles results in a closely viscosity reading for PPcoE/W10L and PPcoE/W20L. The increasing in wollastonite content also increased the peak intensity ratio of oxidised products and a new formation of a tertiary alcohol. This result trend suggested the wollastonite filler was accelerated the TMD of polymer during extrusion. When decreasing in the wollastonite filler size, the composite shows better properties. The rheology of PPcoE/W10S and PPcoE/W20S composites had a higher flow rate reading compare to larger size wollastonite filled composite, the overall amount of degraded products also found reduced, this improvement also detected in thermal stability.

References

- [1] Dogan F 2012 *Polypropylene* InTech. Sd. (Croatia)
- [2] Yu L, Wu T, Chen T, Yang F and Xiang M 2014 *Thermochim. Acta* **578** 43
- [3] Wypych G 2010 *Handb. Fill.* 561
- [4] May P A and Moore J S 2013 *Chem. Soc. Rev.* **42** 7497
- [5] Wang K *et al.* 2012 *Polym. Degrad. Stab.* **97**(8) 1475
- [6] Moghaddam L *et al.* 2011 *Polym. Degrad. Stab.* **96**(4) 455
- [7] Farahanchi A and Sobkowicz M J 2017 *Polym. Degrad. Stab.* **138** 40
- [8] Jose J, Nag A and Nando G B 2010 *J. Polym. Environ.* **18**(3) 155
- [9] Tocháček J, Jančář J, Kalfus J, Zbořilová P and Buráň Z 2008 *Polym. Degrad. Stab.* **93**(4) 770
- [10] Tátraaljai D, Major L, Földes E and Pukánszky B 2014 *Polym. Degrad. Stab.* **102**(1) 33

- [11] Jing X, Yan G, Zhao Y, Wen H and Xu Z 2014 *Polym. Degrad. Stab.* **109** 79
- [12] Ariff Z M, Ariffin A, Rahim N A A and Jikan S S 2012 *InTech* 29
- [13] Rahim N A A, Ariff Z M and Ariffin A 2011 *Pertanika J. Sci. Technol.* **19**(August 2010) 383
- [14] Zhao W *et al.* 1996 *Polym. Degrad. Stab.* **53**(1) 129
- [15] Mastral J F, Berrueco C, Gea M and Ceamanos J 2006 *Polym. Degrad. Stab.* **91**(12) 3330
- [16] Kumanayaka T O, Parthasarathy R and Jollands M 2010 *Polym. Degrad. Stab.* **95**(4) 672
- [17] Kong Q, Hu Y, Song L and Tang Y 2011 *J. Therm. Anal. Calorim.* **104**(3) 1145
- [18] Ojeda T F M, Dalmolin E, Forte M M C, Jacques R J S, Bento F M and Camargo F A O 2009 *Polym. Degrad. Stab.* **94**(6) 965
- [19] Albano C, González J, Ichazo M and Kaiser D 1999 *Polym. Degrad. Stab.* **66**(2) 179
- [20] Gibert J P, Lopez Cuesta J M, Bergeret A and Crespy A 2000 *Polym. Degrad. Stab.* **67**(3) 437
- [21] Bolbukh Y, Kuzema P, Tertykh V and Laguta I 2008 *J. Therm. Anal. Calorim.* **94**(3) 727
- [22] Saw L T, Rahim N A A and Lan D N U 2014 *Malaysian J. Anal. Sci.* **18**(2) 360
- [23] Saw L T, Uy Lan D N, Rahim N A A, Mohd Kahar A W and Viet C X 2015 *Polym. Degrad. Stab.* **111** 32
- [24] Starý Z, Krüchel J, Weck C and Schubert D W 2013 *Compos. Sci. Technol.* **85** 58
- [25] Bellayer S, Tavard E, Duquesne S, Piechaczyk A and Bourbigot S 2009 *Polym. Degrad. Stab.* **94**(5) 797
- [26] Wypych G 2016 *Handb. Fill.* (May 1996) 467