T.R.
EGE UNIVERSITY
Graduate School of Applied and Natural Science

ORIGINAL AND WASTE POLYVINYL BUTYRAL (PVB) / POLYVINYL CHLORIDE (PVC) COMPOSITES WITH NANOADDITIVES AND THE DEGRADATION BEHAVIOUR

MSc THESIS

Cansu KÖROĞLU

Chemical Engineering Department

İzmir
2019

T.R.
EGE UNIVERSITY
Graduate School of Applied and Natural Sciences

ORIGINAL AND WASTE POLYVINYL BUTYRAL (PVB) / POLYVINYL CHLORIDE (PVC) COMPOSITES WITH NANOADDITIVES AND THE DEGRADATION BEHAVIOUR

Cansu KÖROĞLU

Supervisor: Prof. Dr. Serap CESUR

Chemical Engineering Department
Chemical Engineering Second Cycle Programme with Thesis

İzmir
2019

Cansu KÖROĞLU tarafından yüksek lisans tezi olarak sunulan "Original and waste Polyvinyl Butyral (PVB) / Polyvinyl Chloride (PVC) composites with nanoadditives and the degradation behavior (Taze ve atık kaynaklı Polivinil Butiral (PVB) / Polivinil klorür (PVC) nanokatkılı kompozitleri ve bozunma davranışı)" başlıklı bu çalışma EÜ Lisansüstü Eğitim ve Öğretim Yönetmeliği ile EÜ Fen Bilimleri Enstitüsü Eğitim ve Öğretim Yönergesi'nin ilgili hükümleri uyarınca tarafımızdan değerlendirilerek savunmaya değer bulunmuş ve 31 Temmuz 2019 tarihinde yapılan tez savunma sınavında aday oybirliği/~~oyçokluğu~~ ile başarılı bulunmuştur.

Jüri Üyeleri: **İmza**

Jüri Başkanı : **Prof. Dr. Serap CESUR**

Raportör Üye : **Prof. Dr. Mustafa DEMİRCİOĞLU**

Üye : **Doç. Dr. Aylin ZİYLAN**

EGE ÜNİVERSİTESİ FEN BİLİMLERİ ENSTİTÜSÜ

ETİK KURALLARA UYGUNLUK BEYANI

EÜ Lisansüstü Eğitim ve Öğretim Yönetmeliğinin ilgili hükümleri uyarınca Yüksek Lisans Tezi olarak sunduğum "Original and waste Polyvinyl Butyral (PVB) / Polyvinyl Chloride (PVC) composites with nanoadditives and the degradation behavior (Taze ve atık kaynaklı Polivinil Butiral (PVB) / Polivinil klorür (PVC) nanokatkılı kompozitleri ve bozunma davranışı)" başlıklı bu tezin kendi çalışmam olduğunu, sunduğum tüm sonuç, doküman, bilgi ve belgeleri bizzat ve bu tez çalışması kapsamında elde ettiğimi, bu tez çalışmasıyla elde edilmeyen bütün bilgi ve yorumlara atıf yaptığımı ve bunları kaynaklar listesinde usulüne uygun olarak verdiğimi, tez çalışması ve yazımı sırasında patent ve telif haklarını ihlal edici bir davranışımın olmadığını, bu tezin herhangi bir bölümünü bu üniversite veya diğer bir üniversitede başka bir tez çalışması içinde sunmadığımı, bu tezin planlanmasından yazımına kadar bütün safhalarda bilimsel etik kurallarına uygun olarak davrandığımı ve aksinin ortaya çıkması durumunda her türlü yasal sonucu kabul edeceğimi beyan ederim.

31 / 07 / 2019

Cansu KÖROĞLU

ÖZET

TAZE VE ATIK KAYNAKLI POLİVİNİL BUTİRAL (PVB) / POLİVİNİL KLORÜR (PVC) NANOKATKILI KOMPOZİTLERİ VE BOZUNMA DAVRANIŞI

KÖROĞLU, Cansu

Yüksek Lisans Tezi, Kimya Mühendisliği Anabilim Dalı

Tez Danışmanı: Prof. Dr. Serap CESUR

Temmuz 2019, 90 sayfa

Plastikleştirilmiş PVB ve plastikleştirilmiş PVC'den çeşitli derişimlerde Polivinil Butiral (B) / Polivinil klorür (P) filmlerinin hazırlanması ve tanımlanması üzerine çalışılmıştır. PVB'nin en önemli kullanım alanı güvenlik camları olup, geri dönüşüme giren güvenlik camından kazanılan PVB tekrar kullanılabilir ve PVC gibi benzer özellikteki polimerlerle harmanlanabilir. Polimerik filmleri hazırlamak için çift gidiş gelişli W50 ve sürtünme oranı 2:3 olan Brabender yoğurucu cihazı kullanılmıştır. Öncelikle, PVB/PVC filmlerinde yer alan PVB'nin orijinal (OB) ve geri dönüştürülmüş (RB) kaynaklı olmasının etkisi araştırılmıştır. Daha sonra, bu filmlere, iki nano katkı maddesinin (Kil (C) ve Kalsit-$CaCO_3$ (K)) ve bunların oranlarının etkisi değerlendirilmiştir.

Üretilen filmlerin farklı uygulama alanlarında yeni bir ürün olarak değerlendirilebilmesi için kimyasal, mekanik ve termal özellikleri FTIR, Mekanik özellikler, XRD, DSC, TGA, Optik Mikroskop, SEM ve Renk Ölçümü cihazları ile belirlenmiştir. Fourier Dönüşümlü Kızılötesi (FTIR) analizi ile her film için beş farklı pozisyonda fonksiyonel gruplar belirlenmiştir. Filmler oldukça üniform yapıda olup yeni bir fonksiyonel grubun oluşumu gözlenmemiştir. Polimerik kompozit filmlere ağırlıkça %5 Kalsit (K) ilavesi, mekanik özellikler açısından, ağırlıkça %5 kil (C) ilavesinden daha iyi sonuçlar vermiştir. Ancak, orijinal OB90_P10 ve geri dönüşümlü RB90_P10 filmlerine ağırlıkça %0,1 kil eklenmesiyle, en yüksek gerilme mukavemeti ve kopmada uzama değerleri elde edilmiştir. Çünkü az miktarda kil ilavesi, kristallenme, kristal yapısı ve

kristallenme kinetikleri üzerinde bir etkiye sahiptir, bu nedenle bu filmin, ticari olarak kullanılan PVC malzemelerden daha iyi gerilme mukavemetine sahip olduğu görülmektedir. Ayrıca, XRD analizinden görüldüğü gibi ağırlıkça %5 kil ilavesi, kristallik derecesini düşürerek, mekanik özelliklerin azalmasına neden olmuştur. DSC analizi ile orijinal PVB'nin camsı geçiş sıcaklığının (Tg) yaklaşık 18 °C olduğu ve geri dönüşümlü PVB'nin yaklaşık 22 °C olduğu belirlenmiştir. PVC'nin camsı geçiş sıcaklığı -26 °C civarında bulunmuştur. Camsı geçiş sıcaklığı, PVB'ye PVC eklenmesiyle arttığı görülmüştür. Aynı zamanda, tek camsı geçiş sıcaklığı elde edilmesi filmlerin yüksek uyumluluğu olduğunu göstermektedir. Renk ölçüm testinden, polimerik filmlerin renklerinin, ağırlıkça %0,1 ve %1 kil miktarı ilavesi ile değişmediği, fakat ağırlıkça %5 kil miktarı ilavesi ile sarılık değerinin arttığı gözlenmiştir. Filmlerde ağırlıkça %5 Kalsit ilavesi ile renk değişikliği gözlenmemiştir. Polimerik filmlerin termal bozunma davranışı ayrıca TGA termogramları ile analiz edilmiştir. Orijinal PVB, % kül değerine göre diğerlerinden daha düşük değere sahiptir. PVC'nin (P100) % kül miktarı 600 °C'de %12,16 olarak bulundu. % Kül değerlerinin sonuçları OPVB ve RPVB'den daha yüksektir ve orijinal ve geri dönüştürülmüş PVB'ye PVC eklendiğinde, PVC'den daha düşük değer elde edilmiştir. Orijinal ve geri dönüştürülmüş B90_P10 polimerik filmlere ağırlık olarak %0,1 kil ilave edilmesi ile kül değerinin miktarı hafifçe azalır. Kalsiyum karbonat ilavesiyle % kül değerinin sonucu diğer polimerik filmlerden daha yüksektir ve en iyi yanmazlık özelliği elde edilmiştir. Sonuç olarak, az miktarda kil içeren (%0,1) geri dönüştürülmüş PVB / PVC esaslı kompozit filmler orijinal PVC yerine önerilebilir.

Bu çalışma, atık camın neden olduğu çevre sorunlarının çözümüne yönelik bir girişim olup, elde edilen malzemeler döşeme, tutkal üretimi, inşaat mühendisliği gibi farklı uygulamalarda kullanılabilir.

Anahtar kelimeler: polivinil butiral, güvenlik camı, geri dönüşüm, polivinil klorür, kompozit malzeme

ABSTRACT

ORIGINAL AND WASTE POLYVINYL BUTYRAL (PVB) / POLYVINYL CHLORIDE (PVC) COMPOSITES WITH NANOADDITIVES AND THE DEGRADATION BEHAVIOR

KÖROĞLU, Cansu
MSc in Chemical Eng.
Supervisor: Prof. Dr. Serap CESUR

July 2019, 90 pages

The aim of this study is the preparation and characterization of Polyvinyl Butyral (PVB) / Polyvinyl chloride (PVC) blends consisting of plasticized original and waste PVB (B) and plasticized PVC (P) at various concentrations. The common usage area of PVB is safety glass, and waste PVB obtained from recycled safety glass could be utilized again in blends with similar polymers such as PVC. The Brabender kneader device which contained two blungers W50 and a friction ratio of 2:3 was used to prepare polymeric films. The properties of the original and recycled plasticized PVB, used as constituents of the blend was analyzed and compared. The contribution of two additives (Clay (C) and $CaCO_3$ (K)) into the blends was evaluated. The materials obtained from this experiment can be used in various application areas such as flooring, production of glue and in civil engineering.

In order to evaluate the films as a new product for other application areas, their chemical, mechanical, and thermal properties have been characterized by using FTIR, Mechanical properties, XRD, DSC, TGA, Optical Microscopy, SEM and Color Measurement devices. The Fourier Transform InfraRed (FTIR) analysis has been used to determine the film's functional groups in five different positions for each film. The films were uniform and no formation of a new functional group was observed. Although, the mechanical test results show that the addition of 5 wt% $CaCO_3$ (K) into polymeric films have given better results than the addition of 5wt% clay, original OB90_P10 and recycled RB90_P10 composite films which

contained 0.1% (wt/wt) clay (C) showed the highest tensile strength and elongation at break values. As the addition of small amount of clay has an impact on crystallization, crystal structure and crystallization kinetics, this film has better tensile strength than widely used PVC materials. Clay addition of 5 wt% has decreased the degree of crystallinity and has resulted in a decrease at the mechanical properties. The DSC analysis determined that the glass transition temperature (T_g) of original plasticized PVB was almost 18 °C while in recycled PVB it was around 22 °C. The glass transition temperature of PVC was around -26 °C which has a noticeably lower glass transition temperature than PVB. The addition of PVC into PVB polymeric film was found to increase T_g. However, obtaining a single glass transition temperature shows that the films have high compatibility. The color measurement tests have shown that the addition of 0.1 wt% and 1 wt% clay did not affect the color obviously, but yellowness increased with the addition of 5wt% clay. No color change was observed for the films with the addition of 5 wt% of $CaCO_3$. After the fully examined thermal degradation, surface properties and color changes, only recycled PVB/PVC with small amount of clay (0.1 wt%) based polymeric films can be recommended instead of PVC. Thermal degradation behavior of polymeric composite films is also analyzed by TGA thermograms. The original PVB has a smaller value than others with respect to the amount of ash % value. Remaining weight % of PVC (P100) was found as 12.16 % at 600 °C. The results of ash % values are higher than OPVB and RPVB with addition of PVC into original and recycled PVB but, it is lower than PVC. The addition of 0.1 wt% of clay is slightly decreased the amount of ash % value than original and recycled B90_P10 polymeric films. The result of ash content by the addition of calcium carbonate is higher than other polymeric films and the best flammability is obtained.

This study is an important approach to solve environmental problems caused by waste windshield, and the materials obtained can be used in different applications such as flooring, glue production, civil engineering.

Key words: PVB, PVC, clay, calcium carbonate, safety glass, recycling

PREFACE

Thanks to my thesis supervisor Prof. Dr. Serap Cesur, I have acquired the awareness that master's degree education is important for specialize in my profession. She has consistently been an adviser to me in terms of academic and all areas of life throughout my undergraduate and graduate education period. She encouraged me to go on my Erasmus+ journey for my master's degree in education and to expand my perspective. The opportunity to realize my thesis in the Czech Republic is a part of the greatest changes in my life. I have the honor of having a thesis on a topic that is not particularly common in our country by developing me in academic and social fields by living abroad, recognizing different cultures and people. My first encounter with the polymers was in my undergraduate thesis ‘Antimicrobial and Biodegradable Food Packaging Applications of Polycaprolactone/Chitosan Polymeric Composite Films’. In this thesis, since I was working on food packaging, I had the opportunity to do my internship in Korozo packaging. Doing an internship in Korozo Packaging about printing operations has helped me understand that this is the sector of my dream job. By this means for about two years I have been working as a quality control engineer in Bak Packaging, one of the pioneers of the printing sector. My master's education has enabled me to have a different perspective on business life and to fulfill my profession with confidence. I believe that life is always a journey which includes places to be seen and all the memories to be lived. This thesis is the product of my master's degree process, but the processes on the way to reach this product taught me that learning is a long and continuous journey.

İZMİR

31/07/2019

Cansu KÖROĞLU

TABLE OF CONTENTS

Page

ÖZET vi

ABSTRACT ix

PREFACE xi

TABLE OF CONTENTS xiii

TABLE OF FIGURES xv

LIST OF TABLES xix

1. INTRODUCTION 1

2. LITERATURE SURVEY AND THEORETICAL BACKGROUND 4

2.1 Poly (vinyl Butyral) (PVB) and Properties 4

2.2 PVB in Laminated Glass Types 8

2.3 Polyvinyl Chloride (PVC) and Properties 14

2.4 Additives used for Recycled PVB/PVC Composite Film 17

3. EXPERIMENTAL 20

3.1 Materials 20

3.2 Method 20

3.2.1. Preparation of Polymeric Films 20

3.2.2. Analysis Performed 24

4. RESULTS 30

4.1 Film Appearance and Color Values 30

4.2 Chemical Structural Properties 37

4.3 Mechanical Properties 54

4.4 Thermal Properties 64

TABLE OF CONTENTS (continued)

Page

5. CONCLUSION.....81

ACKNOWLEDGEMENT 83

REFERENCES.....84

CURRICULUM VITAE 88

APPENDICES .....

TABLE OF FIGURES

Figures **Pages**

Figure 2.1 Production of Polyvinyl butyral (PVB) with polyvinyl alcohol and butyraldehyde (Butanal) reaction (Mohammadian-Kohola, 2016). 4

Figure 2.2. Laminated glass.....8

Figure 2.3. The steps of processing the most important glass products. 9

Figure 2.4. Components of Laminated Glass. 11

Figure 2.5 The structure of PVC.....14

Figure 2.6 Types of composite structure of polymer-layered silicate clay materials (Rhim and Ng, 2007). 19

Figure 3.1. Experimental procedure of original and recycled PVB polymeric film preparation with PVC, Clay and Calcium Carbonate addition.....21

Figure 3.2. Instrument of FT-IR analysis. 24

Figure 3.3. Steps of determining the mechanical properties. 25

Figure 3.4. Procedure of thermal degradation. 26

Figure 3.5. The L*, a*, and b* coordinate system.....27

Figure 3.6 DSC for the determination of glass transition temperature.....29

Figure 4.1. FT-IR spectra of original PVB (OB100) polymeric film obtained on five different positions and raw material. 37

Figure 4.2. FT-IR spectra of recycled RB90_P10_C0.1 polymeric composite film obtained on five different positions. 38

Figure 4.3. FT-IR spectra of the original plasticized PVB from literature. 38

TABLE OF FIGURES (continued)

Figures **Pages**

Figure 4.4. FT-IR spectra of the original plasticized PVB obtained in this study. 39

Figure 4.5. FT-IR spectra of P100 obtained in this study. 39

Figure 4.6. FT-IR spectra of selected polymeric film samples. 40

Figure 4.7. XRD patterns of clay, original PVB (OB), PVC (P) and the selected composite films. 41

Figure 4.8. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition.....43

Figure 4.9. The surface morphology of OB80_P20_K5 and P100 polymeric films obtained by optical microscopy. 44

Figure 4.10. Effects of original and recycled PVB polymeric films with the blending of PVC and clay on the surface morphology obtained by optical microscopy. 45

Figure 4.11. Effects of different concentrations of PVB, PVC, clay, and $CaCO_3$ on surface morphologies of polymeric films by SEM analysis. 47

Figure 4.12. Chemical element analysis of OPVB. 51

Figure 4.13. Chemical element analysis of OB90_P10. 51

Figure 4.14. Chemical element analysis of OB90_P10_C0.1.....51

Figure 4.15. Chemical element analysis of RB100.....52

Figure 4.16. Chemical element analysis of RB90_P10. 52

Figure 4.17. Chemical element analysis of RB90_P10_C0.1.....52

TABLE OF FIGURES (continued)

Figures **Pages**

Figure 4.18. Chemical element analysis of OB80_P20_K5. 53

Figure 4.19. Chemical element analysis of P100. 53

Figure 4.20. The mechanical test results for the selected polymeric films..... 54

Figure 4.21. Comparison of tensile strength (MPa) of original PVB and recycled PVB in blend with PVC. 56

Figure 4.22. Comparison of elongation at break (%) of original PVB and recycled PVB in blend with PVC. 56

Figure 4.23. Comparison of tensile strength (MPa) of original PVB and recycled PVB in blend with PVC, clay and calcium carbonate. 58

Figure 4.24. Comparison of elongation at break (%) of original PVB and recycled PVB in blend with PVC, clay and calcium carbonate. 59

Figure 4.25. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on mechanical surface morphologies of polymeric films by SEM analysis. 60

Figure 4.26. Color changes of selected polymeric films after thermal degradation. 65

Figure 4.27. Comparison of L*, a* and b* values for the selected samples. 66

Figure 4.28. FT-IR spectra of thermally degraded OB100 at 170 °C for 0-20-40-60 minutes. 67

Figure 4.29. FT-IR spectra of thermally degraded RB100 at 170 °C for 0-20-40-60 minutes. 67

Figure 4.30. FT-IR spectra of thermally degraded P100 at 170 °C for 0-20-40-60 minutes. 68

TABLE OF FIGURES (continued)

Figures **Pages**

Figure 4.31. FT-IR spectra of thermally degraded OB90_P10 at 170 °C for 0-20-40-60 minutes.....68

Figure 4.32. FTIR analysis profiles of thermal degradation for OB95_C5 at 0-20-40-60 minutes at 170 °C. 69

Figure 4.33. FTIR analysis profiles of thermal degradation for OB90_P10_C0.1 at 0-20-40-60 minutes at 170 °C.....69

Figure 4.34. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition at 0 minutes and 60 minutes during thermal degradation.....71

Figure 4.35. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on surface morphologies of selected polymeric films during the thermal degradation by Optical Microscopy.....73

Figure 4.36. Thermograms of selected polymeric polymeric films.....78

Figure 4.37. The amount of ash % at 600 °C.....78

Figure 4.38. DSC profile of selected polymeric composite film with recycled PVB.....80

Figure 4.39. DSC profile of selected polymeric composite film with original PVB. 80

LIST OF TABLES

Tables **Pages**

Table 2.1 Application areas and advantages PVB 6

Table 2.2 Physical properties of PVB (Shark Pellets, 2019). 7

Table 2.3 Properties of Glass Types (Savić. J., et. al., 2013) 11

Table 2.4 Characteristics of the interlayer materials (Castori and Speranzini, 2017) 13

Table 2.5. The properties of rigid and plasticized PVC (Cheremisinoff, P. N., 1998) 15

Table 3.1. The sample codes of the OB/P and RB/P polymeric films with and without additives 23

Table 4.1. Visual appearance of B/P polymeric films without additives 31

Table 4.2. Visual appearance of B/P polymeric films with clay addition 34

Table 4.3. Visual appearance of B/P polymeric films with calcium carbonate addition 36

Table 4.4. Vibrational modes and wavenumbers exhibited by PVC (Ramesha, S., et. al., 2017) 40

Table 4.5. Mechanical properties of polymeric films without nano-additives 55

Table 4.6. Mechanical properties of polymeric composite films with clay addition. 57

Table 4.7. Results of DSC analysis for original and recycled PVB/PVC polymeric films 79

Table 4.1. Visual appearance of B/P polymeric films without additives (Continued)

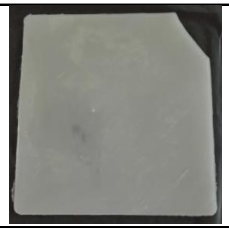
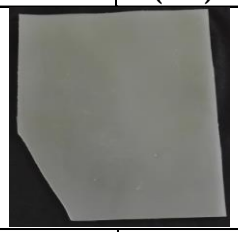
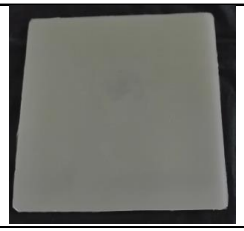
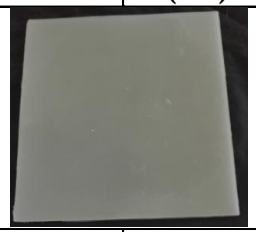
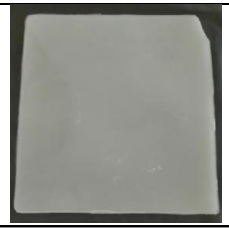
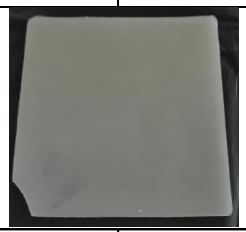
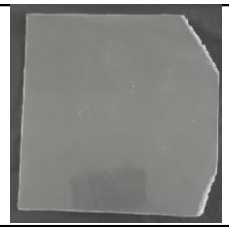
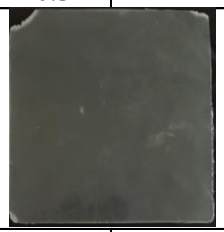
		Original B				Recycled B			
		Sample 1	Sample 2			Sample 1	Sample 2		
B (wt%)	P (wt%)	Weight 1 (g)	Weight 2 (g)	Average Thickness (mm)	Color Values	Weight 1 (g)	Weight 2 (g)	Average Thickness (mm)	Color Values
20	80				L*: 49.04 a*: -0.01 b*: 1.63				L*: 49.33 a*: -0.11 b*: 2.03
		20.3	20.3	"		20.1	20.0	"	
10	90				48.40 1.12 1.53				46.83 0.04 0.83
		21.0	20.5	"		20.5	20.6	"	
0	100				38.38 1.14 3.03				
		20.6	20.0	"					

Table 4.2. Visual appearance of B/P polymeric films with clay addition

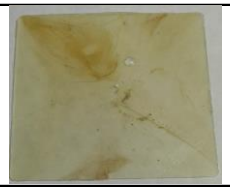
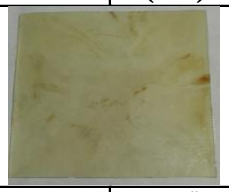
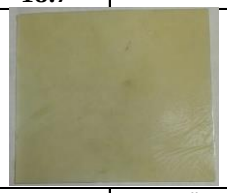
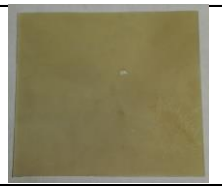
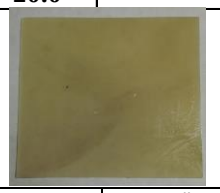
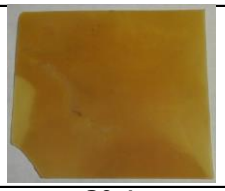
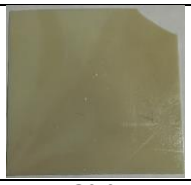
			Original B				Recycled B			
			Sample 1	Sample 2		Colour Values	Sample 1	Sample 2		Colour Values
B (wt%)	P (wt%)	C (wt%)	Weight 1 (g)	Weight 2 (g)	Average Thickness (mm)		Weight 1 (g)	Weight 2 (g)	Average Thickness (mm)	
95	0	5				L*: 37.31 a*: 0.15 b*: 3.51				L*: 37.77 a*: 0.50 b*: 4.48
			19.0	19.3	“		19.8	19.8	“	
90	10	0.1				41.58 0.71 3.75				42.01 0.22 3.97
			18.7	18.7	“		20.5	20.0	“	
90	10	1				41.53 0.74 5.59				44.23 1.28 7.13
			19.6	19.9	“		19.7	19.5	“	
80	20	5				45.09 3.50 13.87				45.34 5.66 14.89
			20.5	20.2	“		20.4	20.4	“	

Table 4.2. Visual appearance of B/P polymeric films with clay addition (Continued)

			Original PVB				Recycled PVB			
			Sample 1	Sample 2			Sample 1	Sample 2		
B (wt%)	P (wt%)	Clay wt%	Weight 1 (g)	Weight 2 (g)	Average Thickness (mm)	Colour Values	Weight 1 (g)	Weight 2 (g)	Average Thickness (mm)	Colour Values
60	40	5				L*: 45.37 a*: 5.06 b*: 15.18				L*: 41.93 a*: 9.29 b*: 17.38
			19.7	19.6	0.05		20.0	20.5	"	
40	60	5				43.94 11.02 22.08				45.04 4.74 16.80
			20.3	20.5	"		21.3	21.6	"	
20	80	5				41.23 9.38 18.24				40.16 4.90 12.86
			20.1	20.0	"		20.2	20.2	"	
0	95	5				35.62 1.70 5.95				
			19.6	19.4	"					

Table 4.3. Visual appearance of B/P polymeric films with calcium carbonate addition

B wt%	P wt%	K wt%	Sample 1	Sample 2		Colour Values
			Weight 1 (g)	Weight 2 (g)	Average Thickness (mm)	
95	0	5				L*: 63.59 a*: -3.58 b*: -1.28
			19.0	18.4	"	
RPVB 95	0	5				64.40 -1.32 3.91
			20.0	20.1	"	
80	20	5				69.71 -1.68 1.84
			20.2	20.9	"	
60	40	5				69.80 -0.84 3.41
			19.5	19.8	"	
40	60	5				68.97 -1.45 2.35
			19.8	20.0	"	
20	80	5				62.32 -1.48 1.11
			20.7	20.6	"	

4.2 Chemical Structural Properties

¾ Fourier Transform Infrared (FT-IR)

The Fourier Transform Infra-Red (FTIR) analysis has been used to determine the functional groups polymeric films at five different positions for each film. FTIR spectra have been used to check the uniformity of the film as can be seen they are very uniform and no formation of a new functional group is observed for all polymeric films as shown in Figures 4.1 and 4.2 for original PVB and recycled RB90_P10_C0.1 polymeric film, respectively. The functional groups of the polymer depend on the plasticizers used. The functional groups of the original and recycled films are the same.

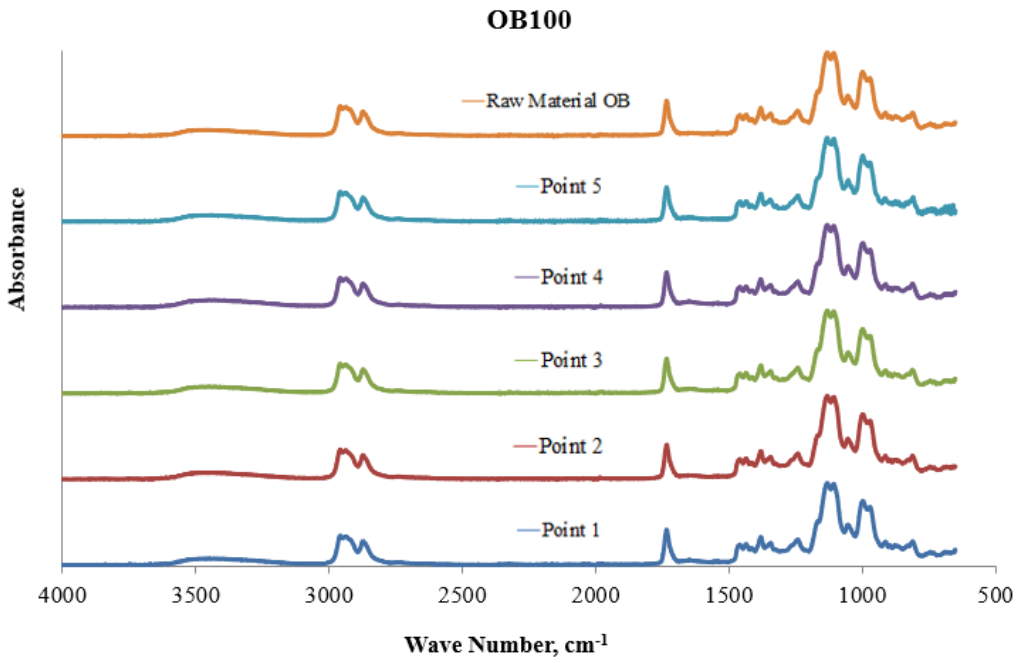


Figure 4.1. FT-IR spectra of original PVB (OB100) polymeric film obtained on five different positions and raw material.

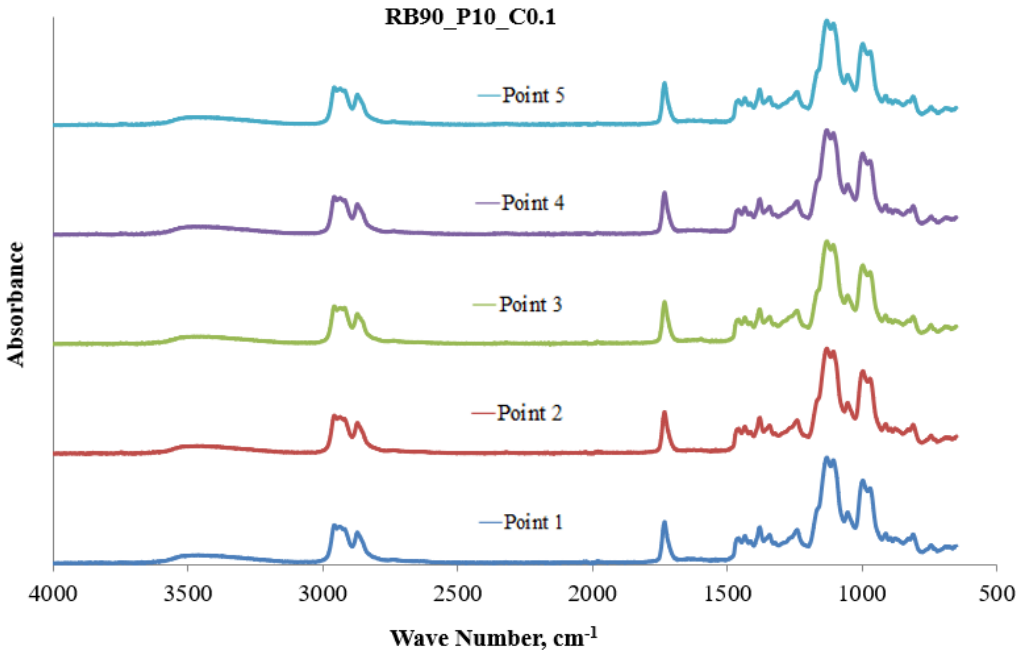


Figure 4.2. FT-IR spectra of recycled RB90_P10_C0.1 polymeric composite film obtained on five different positions.

As can be seen in Figures 4.3 and 4.4, FT-IR spectra of original plasticized PVB obtained from literature and in this study are really consistent.

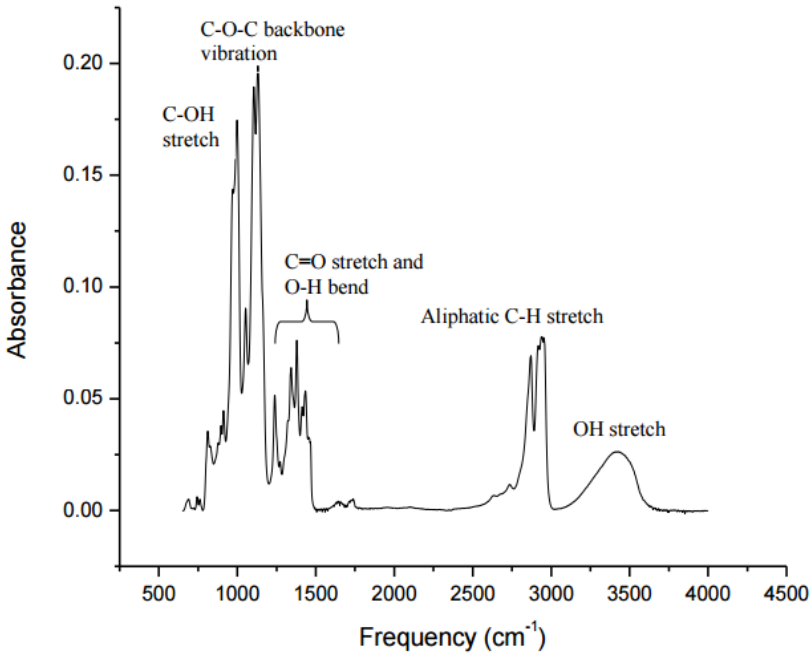


Figure 4.3. FT-IR spectra of the original plasticized PVB from literature.

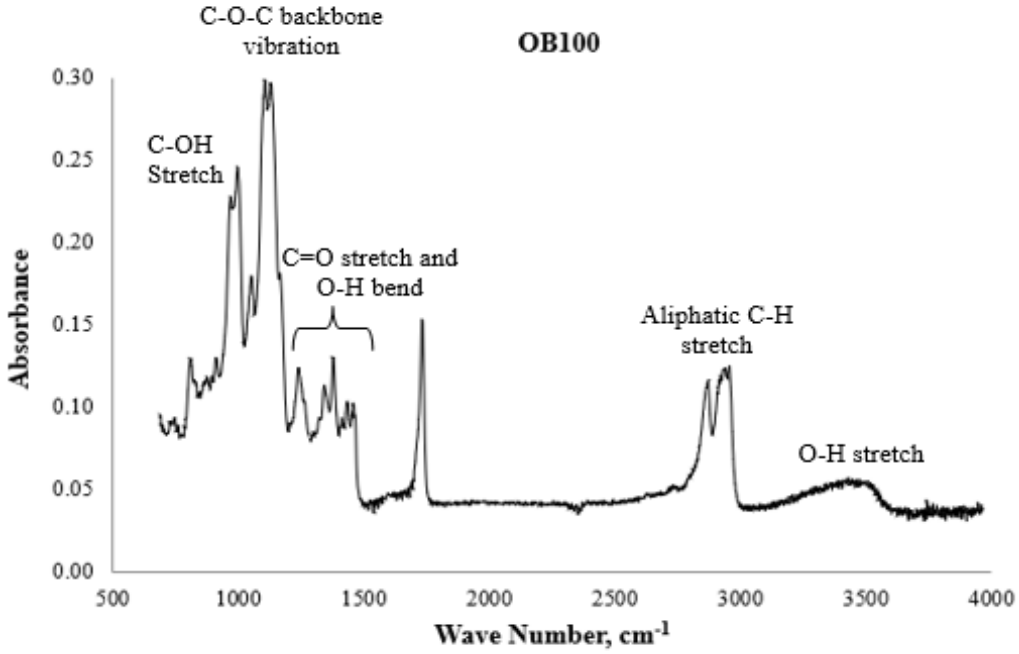


Figure 4.4. FT-IR spectra of the original plasticized PVB obtained in this study.

Figure 4.5 was obtained in this study for the plasticized PVC and Table 4.4 is taken from the literature that is much consistent. They are also in agreement.

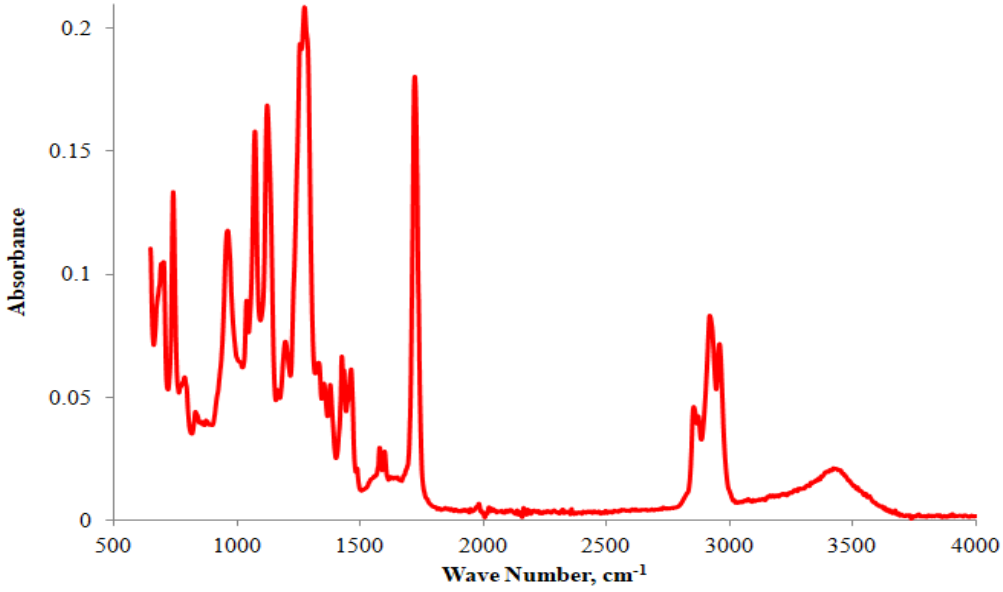


Figure 4.5. FT-IR spectra of P100 obtained in this study.

Table 4.4. Vibrational modes and wavenumbers exhibited by PVC (Ramesha, S., et. al., 2017)

Wavenumber (cm^{-1})	**assignment**
2970	Stretching C-H of CHCl
2912	Stretching C-H of CH_2
1435 and 1427	Deformation (Wagg) CH_2
1331 and 1255	Deformation C-H of CHCl
1099	Stretching C-C
966	Rocking CH_2
692, 637, and 616	Stretching C-Cl

Figure 4.6 gives the FT-IR spectra of the original and recycled PVB and PVC films. The profiles are very similar to each other. The film samples are uniform and no formation of a new functional group is observed.

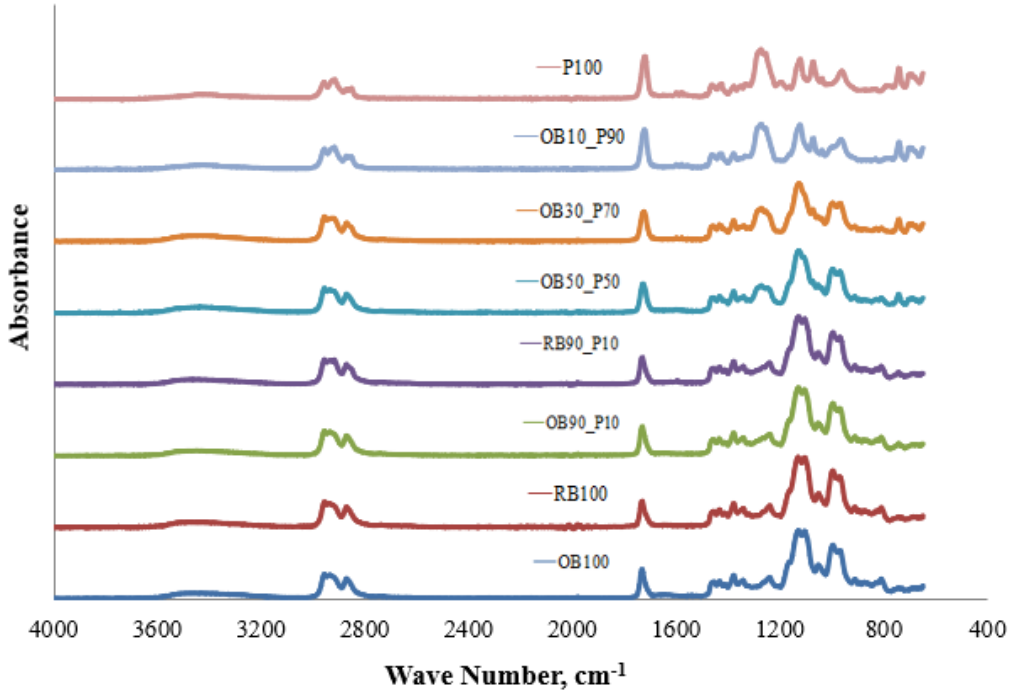


Figure 4.6. FT-IR spectra of selected polymeric film samples.

¾ X-Ray Diffraction (XRD) Analysis

Crystal structures of all original PVB and recycled PVB in blend with PVC, Clay and Calcium Carbonate polymeric films obtained are identified by XRD analysis. In Figure 4.7, the XRD analysis of polymeric films and nano-clay (Closite 30B) contained polymeri F ILOPV FDQ EH VHHQ &OD\ KD degrees at 4.7. The peak of clay is disappearing in the polymeric composite films meaning that clay is intercalated and fully exfoliated in the polymer matrix.

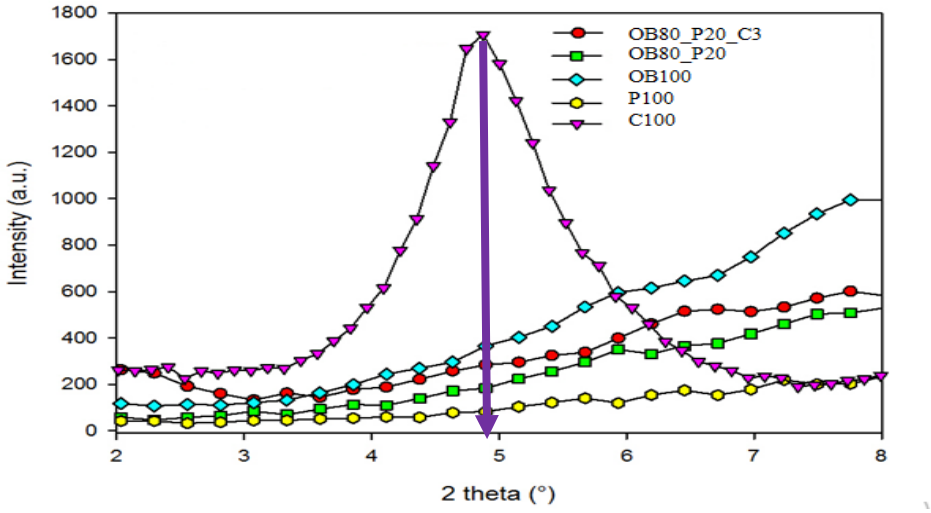


Figure 4.7. XRD patterns of clay, original PVB (OB), PVC (P) and the selected composite films.

IQ)LJXUH WKH PD[LPXP SHDN ZDV REVHUY polymeric films which are the characteristic peaks of PVB and PVC. The original and recycled PVB patterns are almost the same and this shows that recycled PVB has similar degree of crystallinity properties like original PVB. The original and recycled PVB has the highest degree of crystallinity. The degree of crystallinity is decreased slightly with the addition of PVC into original and recycled PVB.

The clay addition of 0.1 wt% amount into original and recycled B90_P10 polymeric film has a small effect on the intensity than original and recycled B90_P10 polymeric films. As the addition of a small amount of clay has an impact on crystallization, crystal structure, and crystallization kinetics. Clay addition of 5 wt% has decreased the degree of crystallinity and caused the mechanical properties decreased that will be discussed in section 4.3.

It can also be seen in Figure 4.8 that the addition of calcium carbonate has different peak positions than the addition of clay because of the chemical structure of calcium carbonate.

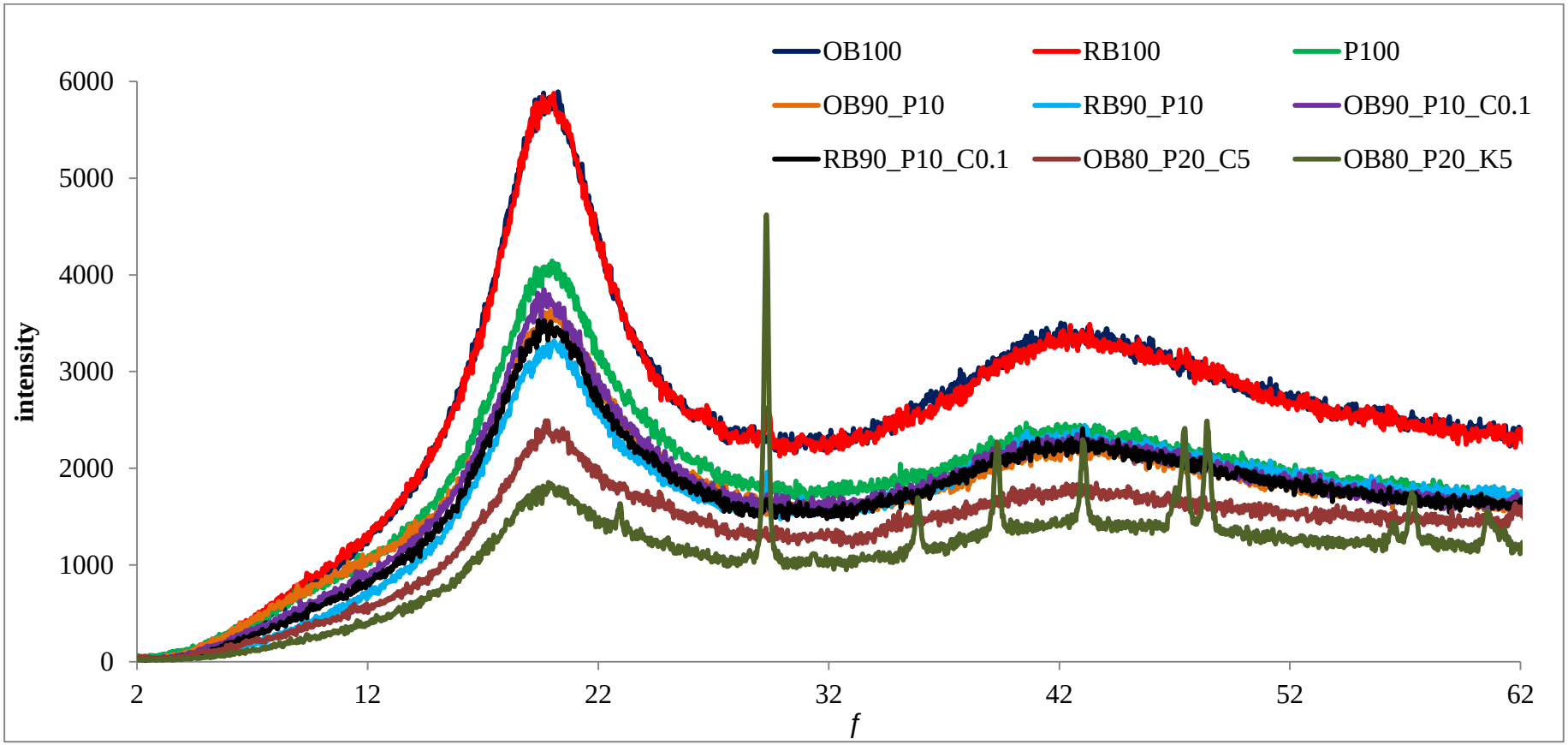


Figure 4.8. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition.

¾ Optical Microscopy

Surface morphology of the original/recycled PVB and PVC with and without nano-additives films was examined by optical microscopy and all results are given in Figure A1 in Appendix Section. Figure 4.9 and 4.10 show the surface morphology of selected polymeric films. Surface morphology of recycled PVB shows more heterogeneous structure than the original PVB because recycled PVB has several impurities. The surface morphology is a bit heterogeneous with the addition of 10 wt% PVC into PVB. The addition of 0.1 wt% clay has similar behavior at surface morphology. PVC, Clay, and Calcium carbonate are dispersed as smaller particles into PVB. The surface morphology of plasticized PVC is slightly heterogeneous than original and recycled plasticized PVB. It can also be said that surface morphology of the all obtained polymeric films with recycled PVB show more heterogeneous structure than the original PVB.

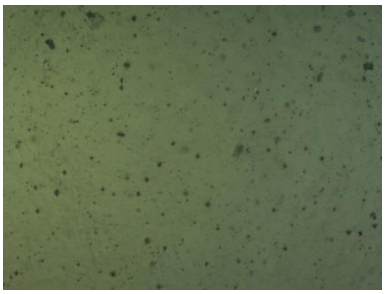
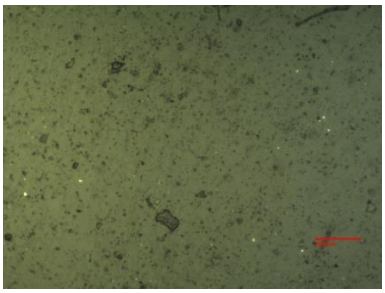
Sample Name	
OB80_P20_K5	
P100	

Figure 4.9. The surface morphology of OB80_P20_K5 and P100 polymeric films obtained by optical microscopy.

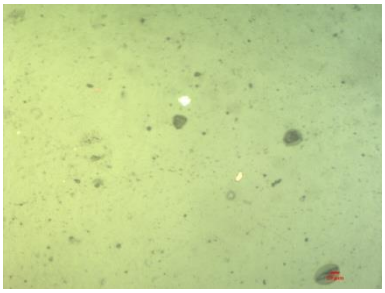
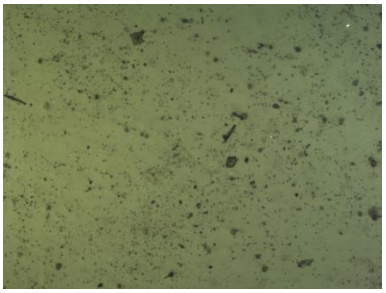
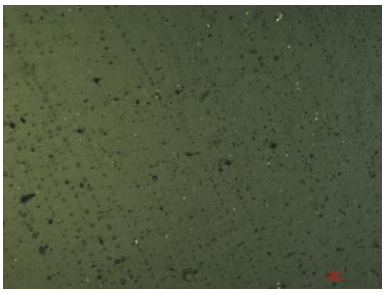
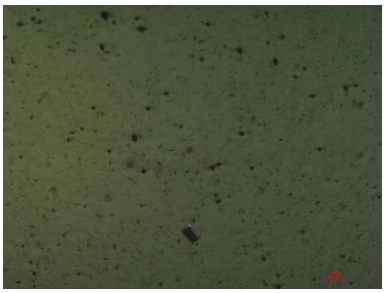
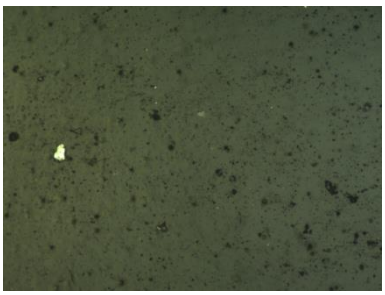
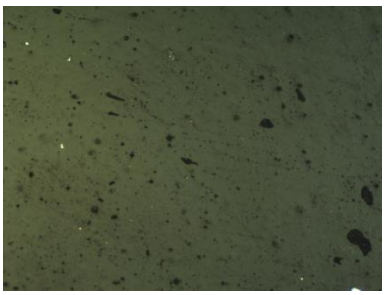
Sample Name	Original PVB	Recycled PVB
B100		
B90_P10		
B90_P10_C0.1		

Figure 4.10. Effects of original and recycled PVB polymeric films with the blending of PVC and clay on the surface morphology obtained by optical microscopy.

¾ **Scanning Electron Microscope** (**SEM)**

Surface morphology of the plasticized PVB and plasticized PVC polymeric films was investigated by SEM and results are given in Figure 4.11 for the selected polymeric films. It can be seen that there occur two separate interfaces with the addition of 10 wt% PVC into the original PVB. The polymeric film with the same amount of RPVB shows opposite behavior and PVB and PVC blend is very homogenous because of good compatibility. When 0.1 wt% clay is added into RB90_P10 polymeric films, well-dispersed surface morphology is obtained. The addition of 5 wt% calcium carbonate affects the surface morphology negatively than the addition of 0.1 wt% clay.

The results of chemical element analyses of selected polymeric films are given in Figure 4.12 ±4.19. According to EDS analyses, it is seen from Figures 4.12, and 4.15 that recycled PVB has small amount Si element which might be obtained during the recycling process from SiO_2 in glass and it is not in the original PVB. From Figures 4.13, and 4.16, 10 wt% PVC added into original and recycled PVB films have Cl element because of PVC structure. Also, OB90_P10 has unexpected different element compositions (Al, Si, K, Ca, Fe) which might be obtained from the plasticizer used or the contamination of the film. 0.1 wt% clay added into OB90_P10 polymeric film shows Na, Al, Si, and Cl elements. It can be seen from Figure 4.18 that the calcium carbonate addition into OB80_P20 polymeric film shows Ca element as expected.

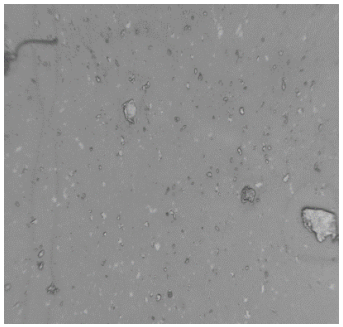
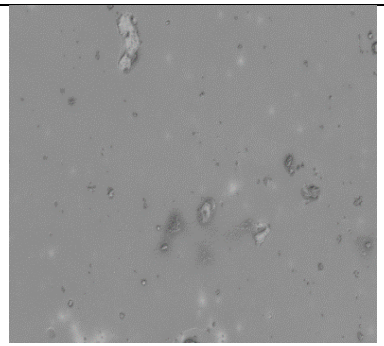
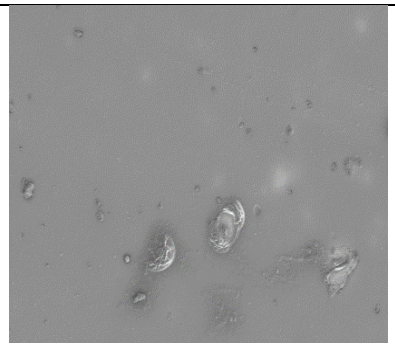
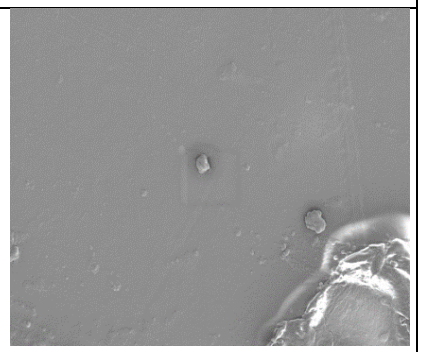
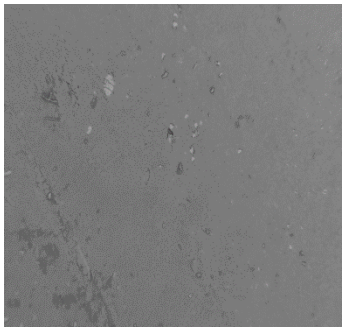
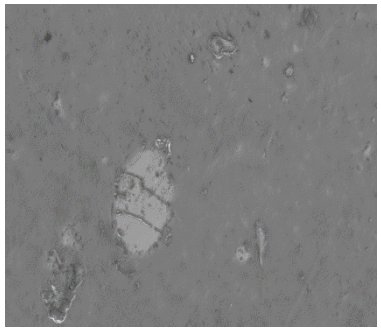
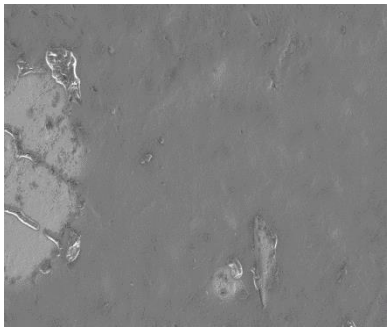
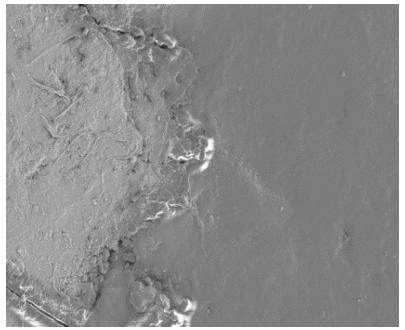
Film	100x	500x	1000x	5000x
OB100				
OB90_P10				

Figure 4.11. Effects of different concentrations of PVB, PVC, clay, and $CaCO_3$ on surface morphologies of polymeric films by SEM analysis.

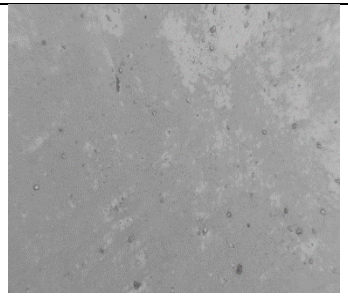
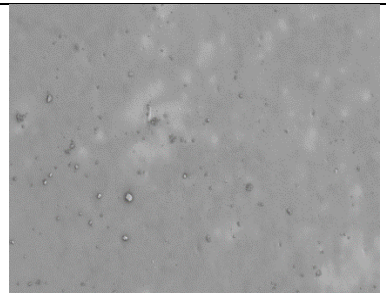
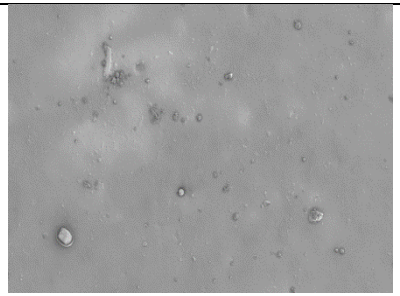
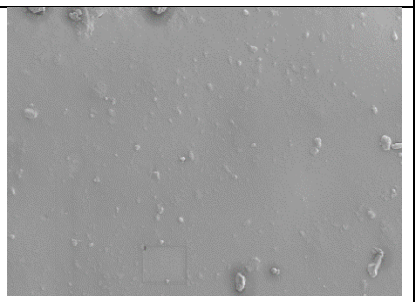
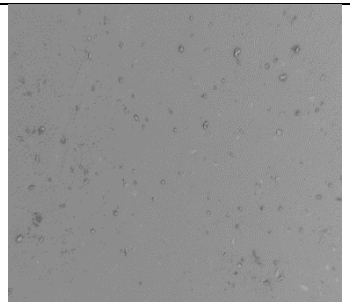
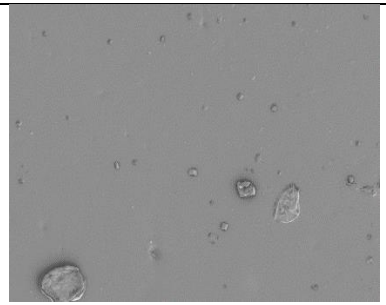
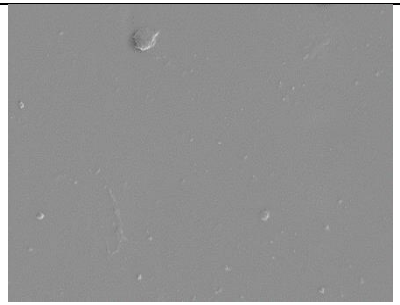
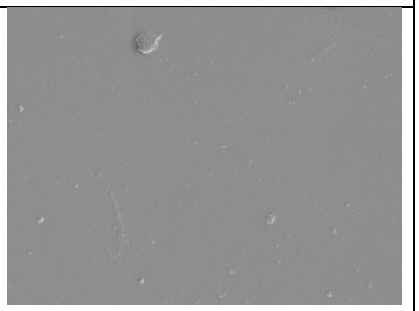
Film	100x	500x	1000x	5000x
OB90_P10_C0.1				
RB100				

Figure 4.11. Effects of different concentrations of PVB, PVC, clay, and $CaCO_3$ on surface morphologies of polymeric films by SEM analysis (Continued).

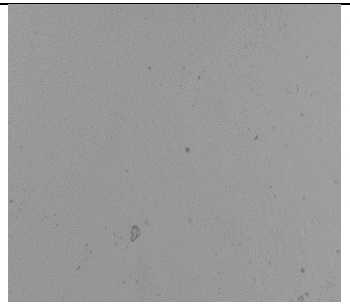
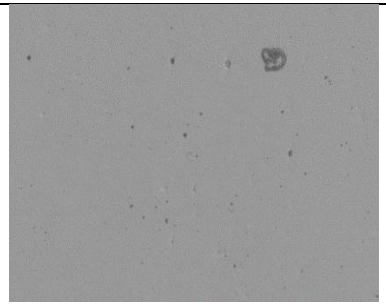
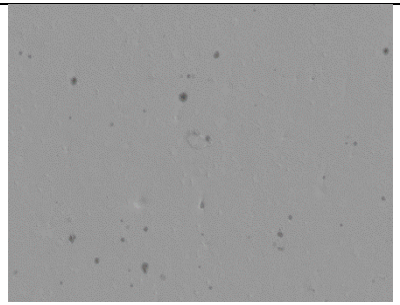
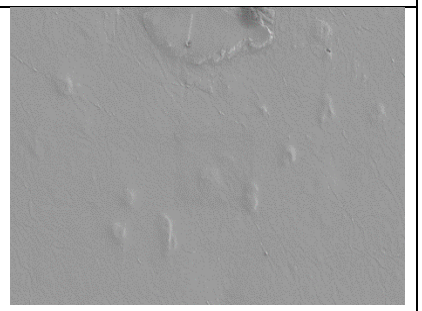
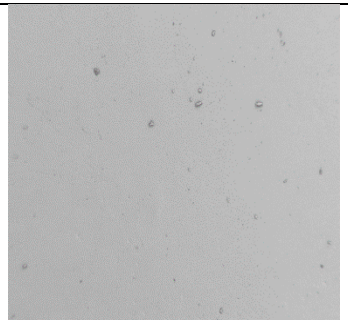
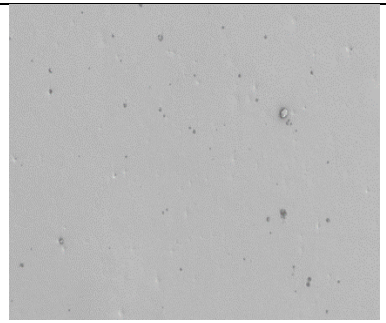
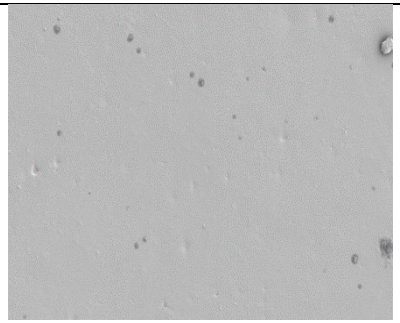
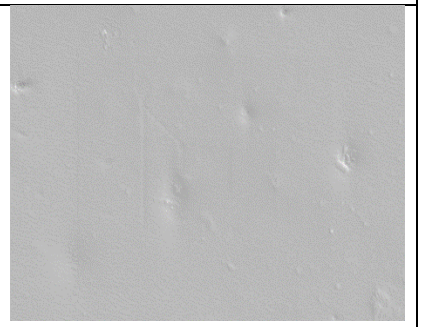
Film	100x	500x	1000x	5000x
RB90_P10				
RB90_P10_C0.1				

Figure 4.11. Effects of different concentrations of PVB, PVC, clay, and $CaCO_3$ on surface morphologies of polymeric films by SEM analysis (Continued).

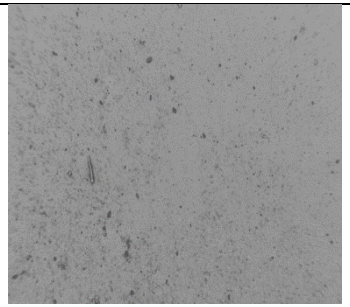
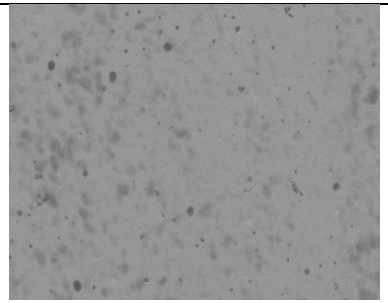
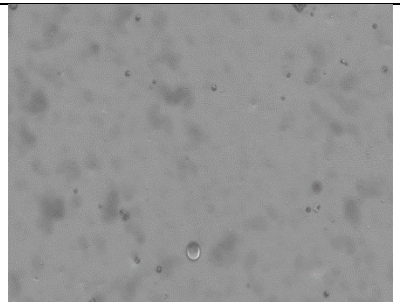
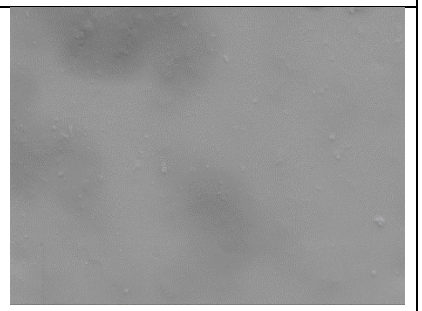
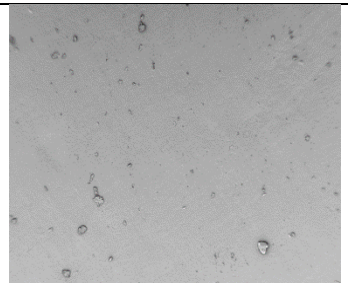
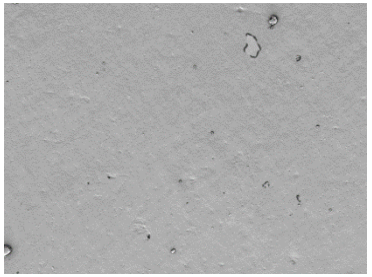
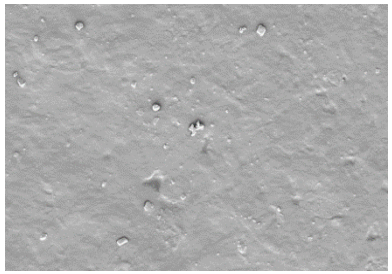
Film	100x	500x	1000x	5000x
OB80_P20_K5				
P100				

Figure 4.11. Effects of different concentrations of PVB, PVC, clay, and $CaCO_3$ on surface morphologies of polymeric films by SEM analysis (Continued).

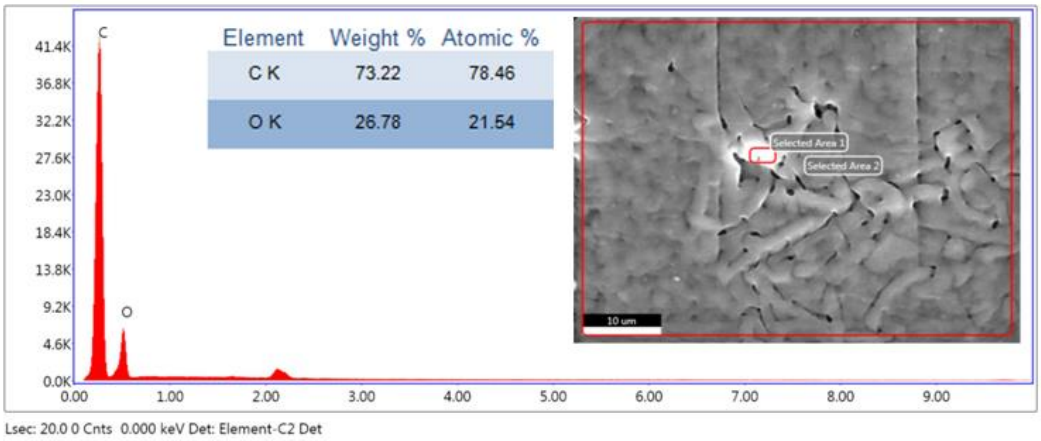


Figure 4.12. Chemical element analysis of OPVB.

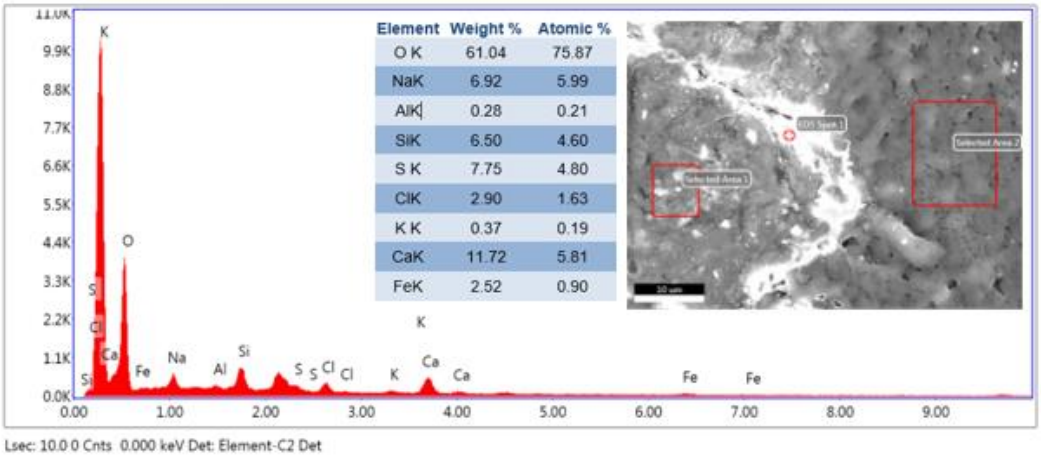


Figure 4.13. Chemical element analysis of OB90_P10.

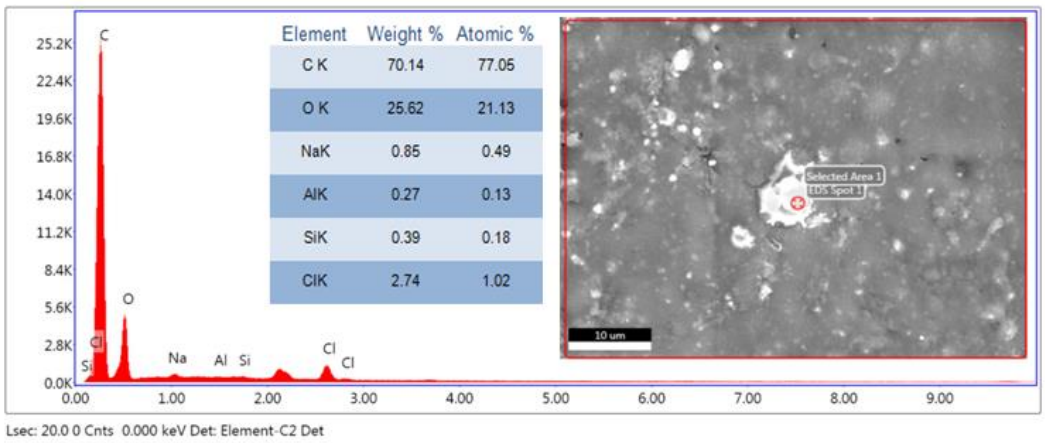


Figure 4.14. Chemical element analysis of OB90_P10_C0.1.

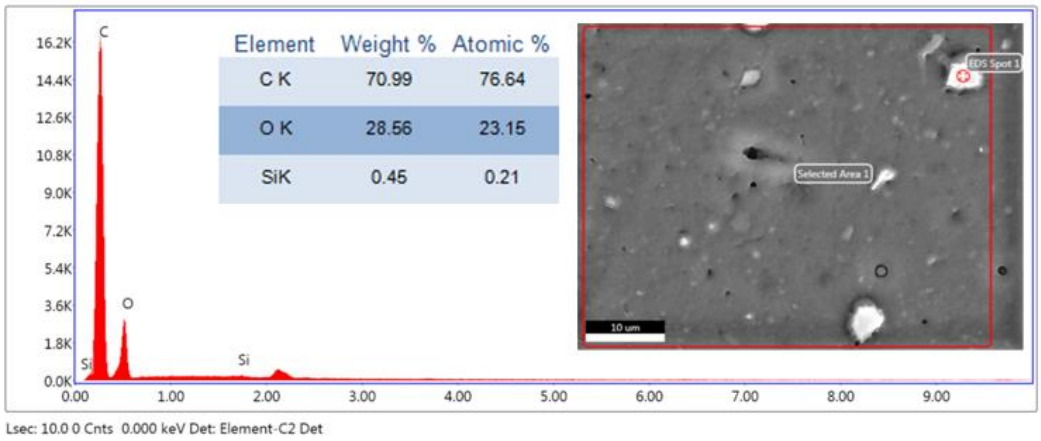


Figure 4.15. Chemical element analysis of RB100.

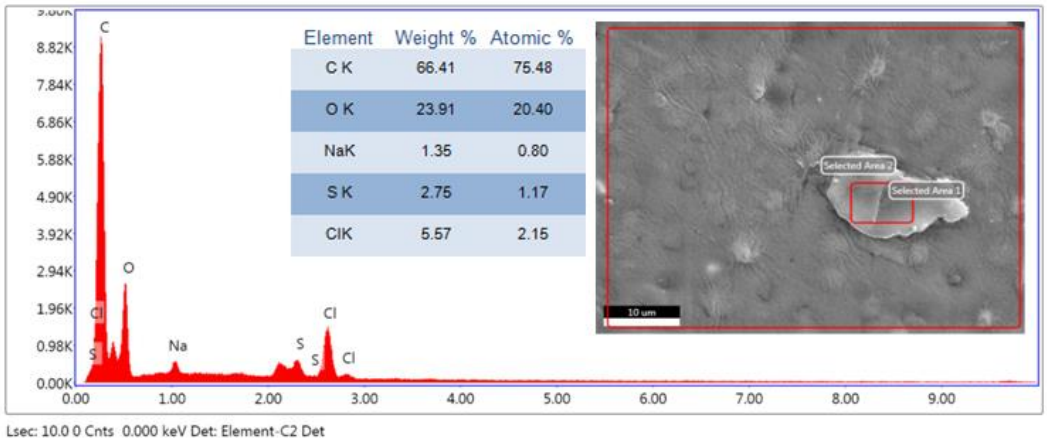


Figure 4.16. Chemical element analysis of RB90_P10.

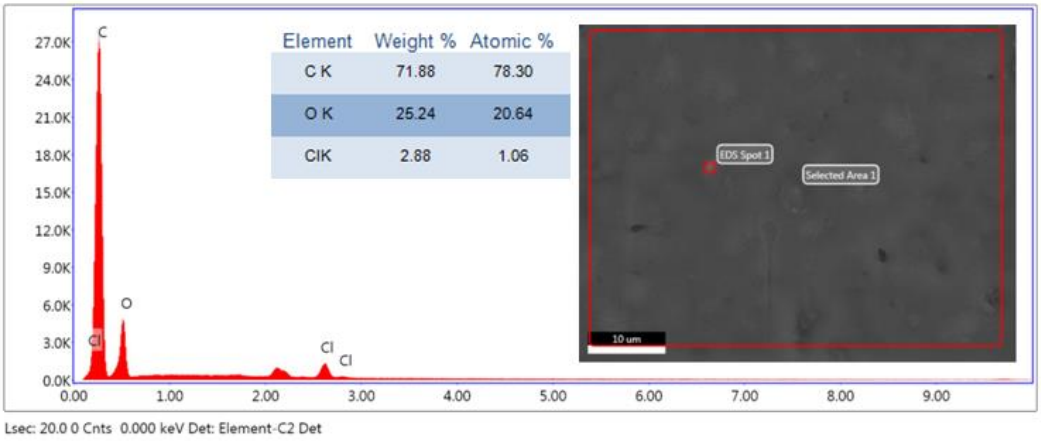


Figure 4.17. Chemical element analysis of RB90_P10_C0.1.

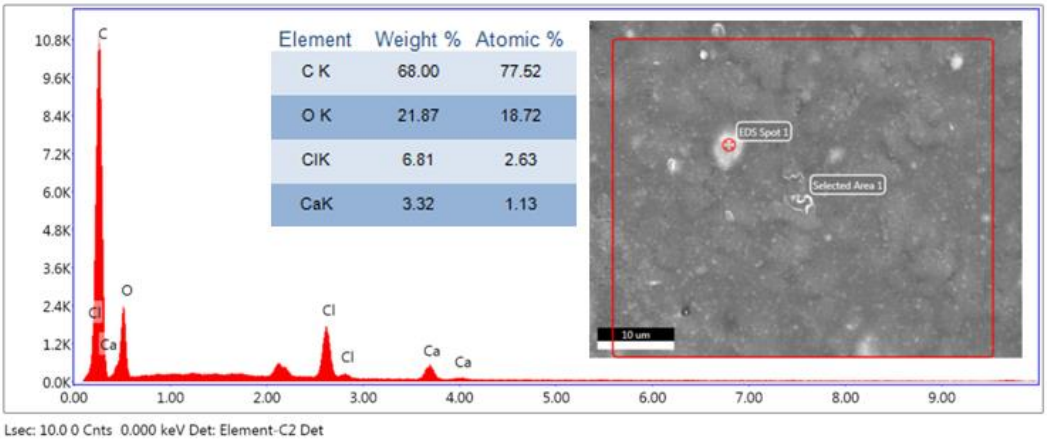


Figure 4.18. Chemical element analysis of OB80_P20_K5.

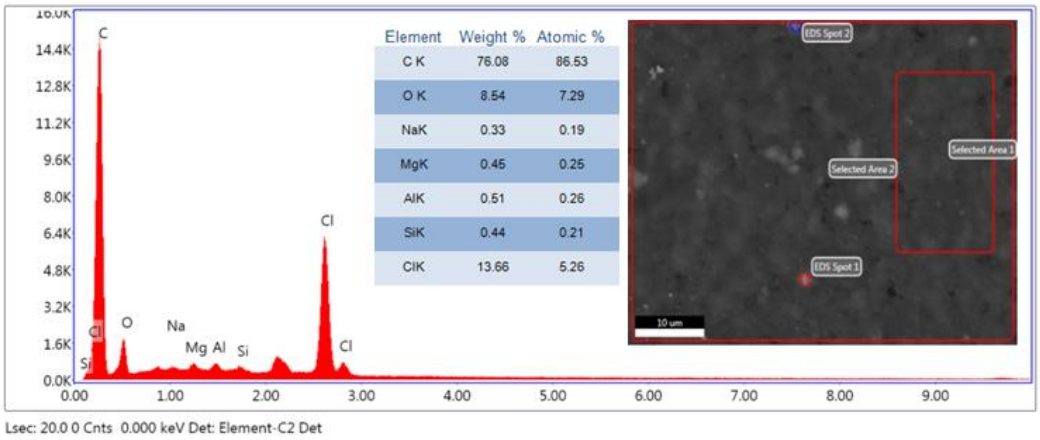


Figure 4.19. Chemical element analysis of P100.

4.3 Mechanical Properties

For each polymeric film, as can be seen in Figure 4.20, the mechanical tests have been repeated six times on six different locations to take an average of mechanical values.

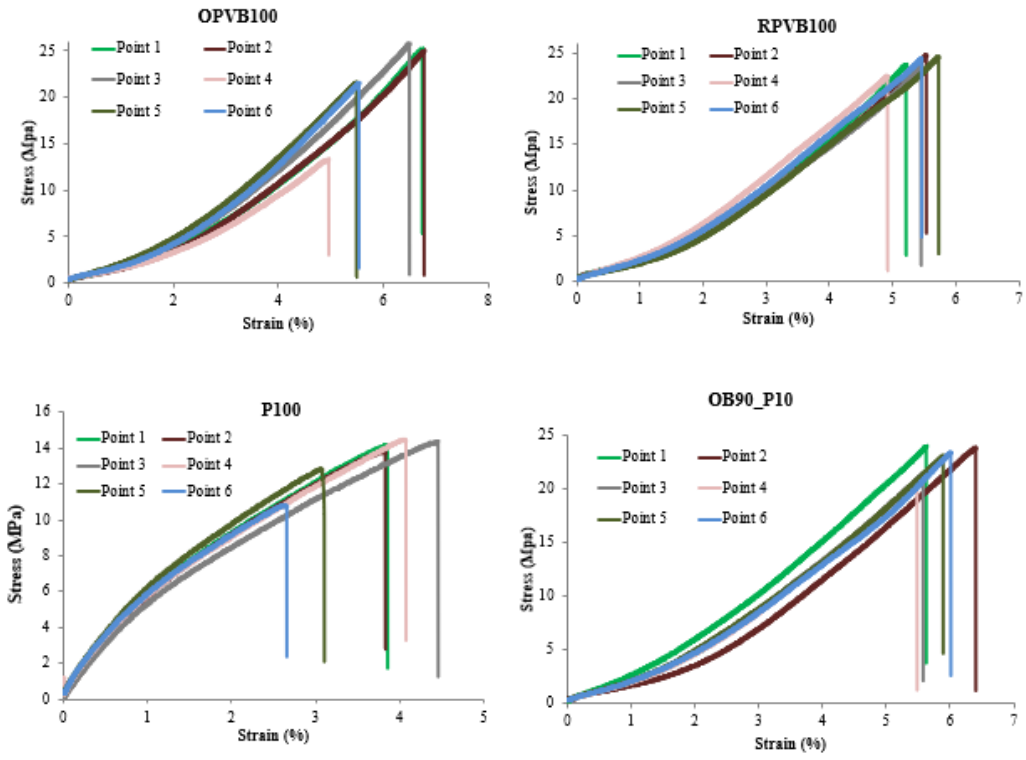


Figure 4.20. The mechanical test results for the selected polymeric films.

First without nano-additives, main polymers as original and recycled plasticized PVB and PVC films have been prepared, and from the mechanical test results obtained, the films that give the good mechanical results have been selected and the nano-additives have been added. For original and recycled PVB, the profiles are almost the same and for the recycled PVB, they are more uniform. The mechanical results are given in Table 4.8. The mechanical property of recycled PVB is slightly less than the original PVB. This proves that the recycled PVB sheets possess physical properties very similar to those of the extruded material before the lamination process, but only the elongation slightly decreases because of the impurities.

On the other hand, it can be further seen from Table 4.5 that the mechanical properties of plasticized PVC were improved with the addition of plasticized

original or recycled PVB after 70 wt% PVB. The obtained polymeric films can be suggested to be used instead of the original plasticized PVC. Moreover, the mechanical properties of the plasticized PVC decrease with the addition of between 10 and 60 wt% original and recycled PVB. The results can be compared in Figures 4.21 and 4.22.

Table 4.5. Mechanical properties of polymeric films without nano-additives

	Tensile Strength (MPa)		**Elongation at break (%)**	
Sample Code	**Original PVB**	**Recycled PVB**	**Original PVB**	**Recycled PVB**
B100	“	“	“	“
B90_P10	“	“	“	“
B80_P20	“	“	“	“
B70_P30	“	“	“	“
B60_P40	“	“	“	“
B50_P50	“	“	“	“
B40_P60	“	“	“	“
B30_P70	“	“	“	“
B20_P80	“	“	“	“
B10_P90	“	“	“	“
P100	“	“	“	“

Results of mechanical tests for polymeric films with the increasing of PVC into the polymeric film is getting worse, actually, original and recycled PVB have very good mechanical properties and the mechanical properties are deteriorating with increasing of the addition of plasticized PVC and original PVC are slightly better than those. It can be concluded that this deterioration is because of the in compatibility of the two different polymers when they are combined into the polymeric film.

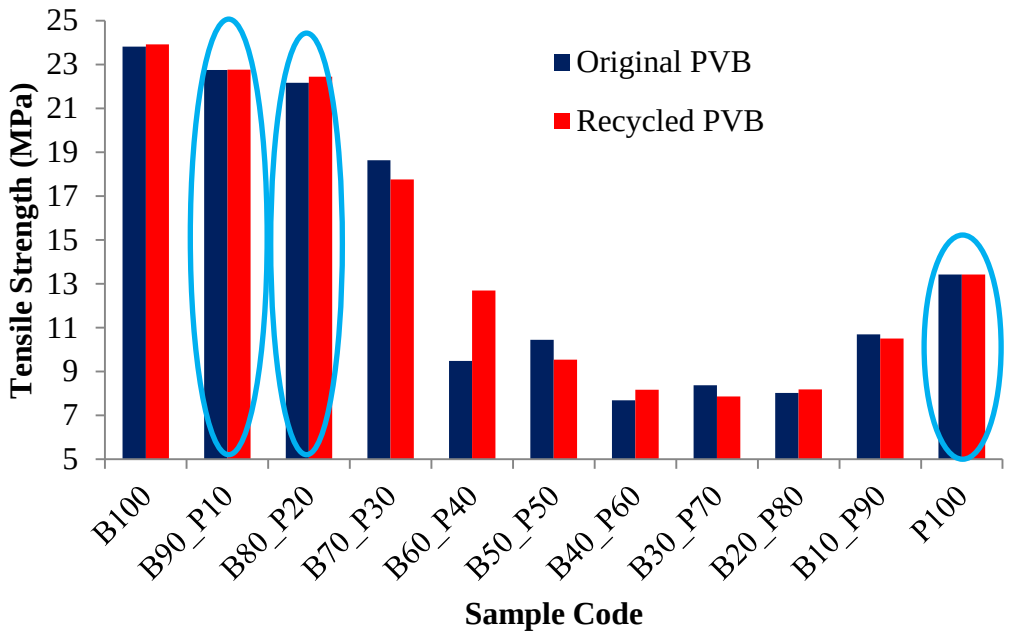


Figure 4.21. Comparison of tensile strength (MPa) of original PVB and recycled PVB in blend with PVC.

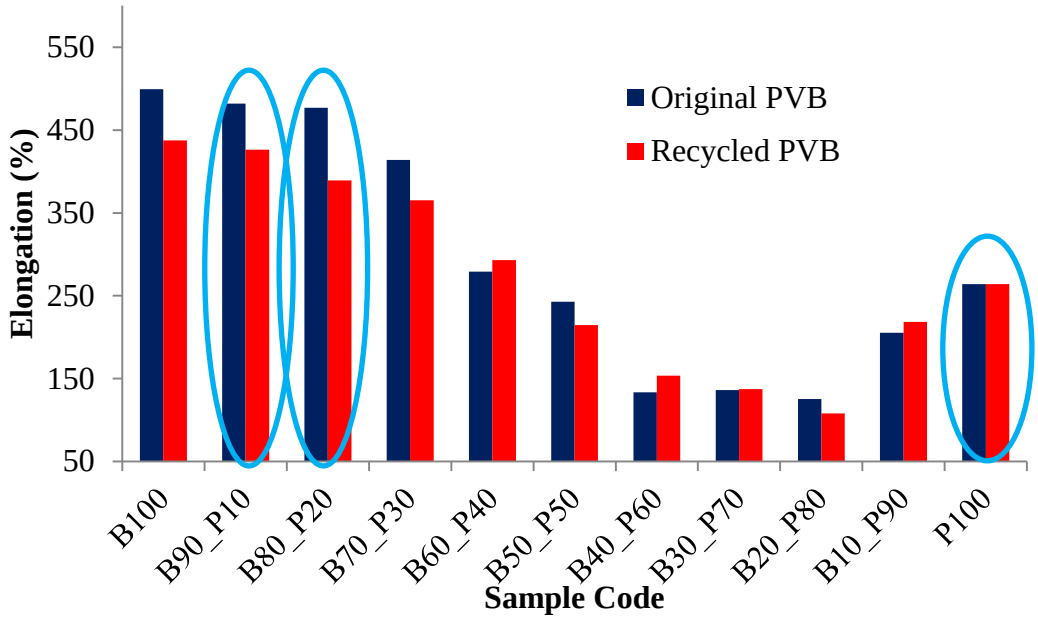


Figure 4.22. Comparison of elongation at break (%) of original PVB and recycled PVB in blend with PVC.

Addition of 10 wt% PVC and 0.1 wt% clay into original PVB and recycled PVB have the best mechanical properties than other polymeric films as shown in Table 4.6 because small amount of clay influences the crystallization kinetics. But, no significant improvement in the mechanical properties of the 5 wt% clay addition has been observed as seen in Table 4.6.

Table 4.6. Mechanical properties of polymeric composite films with clay addition.

	Tensile Strength (MPa)		**Elongation at break (%)**	
Sample Code	**Original PVB**	**Recycled PVB**	**Original PVB**	**Recycled PVB**
B100	"	"	"	"
B95_C5	"	"	"	"
B90_P10	"	"	"	"
B90_P10_C0.1	"	"	"	"
B90_P10_C1	"	"	"	"
B80_P20	"	"	"	"
B80_P20_C5	"	"	"	"
B60_P40	"	"	"	"
B60_P40_C5	"	"	"	"
B40_P60	"	"	"	"
B40_P60_C5	"	"	"	"
B20_P80	"	"	"	"
B20_P80_C5	"	"	"	"
P95_C5	"	"	"	"
P100	"	"	"	"

Comparison of mechanical results of original PVB and recycled PVB in blend with PVC, clay and calcium carbonate can be seen in Figures 4.23 and 4.24. Although, the addition of 5 wt% $CaCO_3$ (K) into polymeric films gave better results than addition of 5wt% clay, but 0.1% (wt/wt) clay (C) added original OB90_P10 and recycled RB90_P10 films showed the highest tensile strength and elongation at break values. Because, the addition of small amount of clay has an impact on the crystallization, crystal structure and crystallization kinetics, therefore, this film has better tensile strength than commercially used PVC materials.

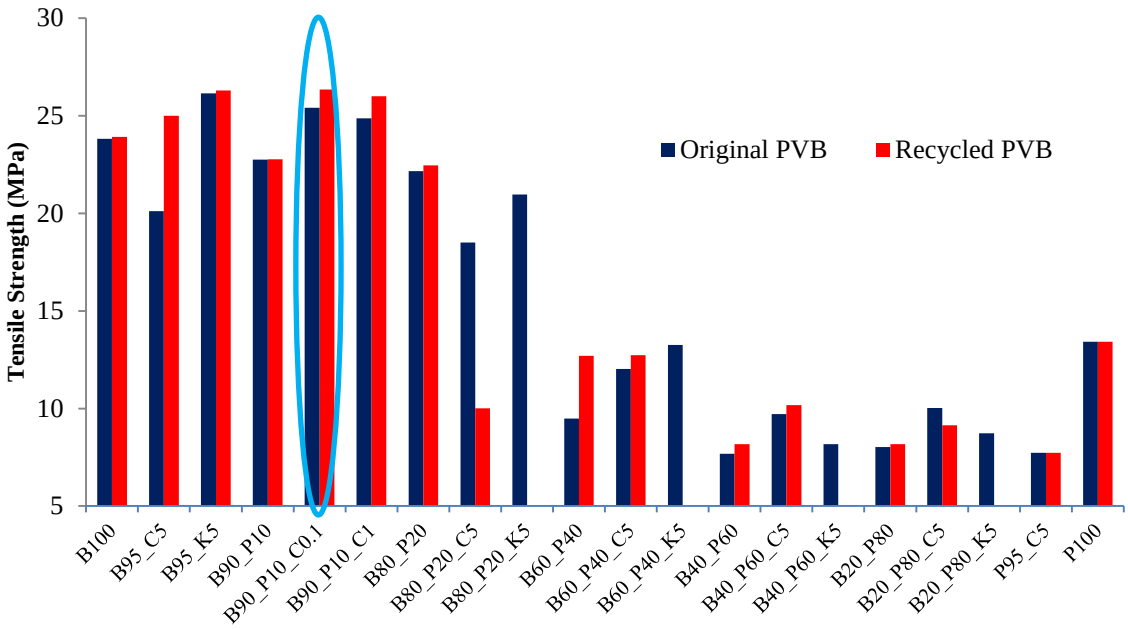


Figure 4.23. Comparison of tensile strength (MPa) of original PVB and recycled PVB in blend with PVC, clay and calcium carbonate.

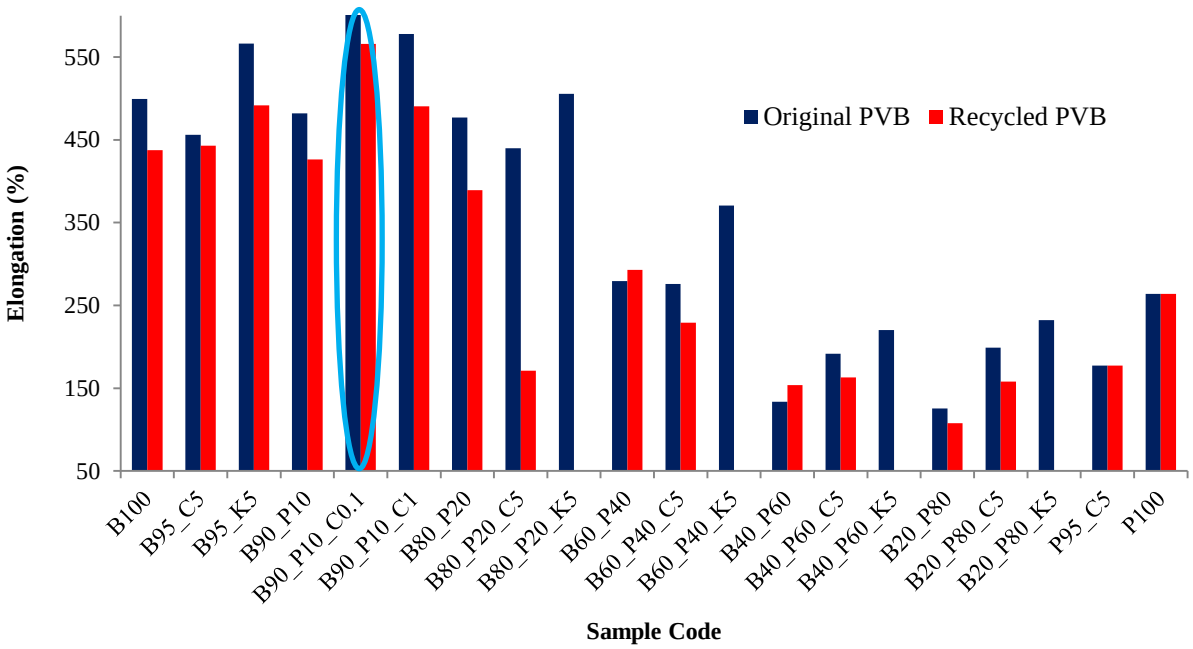


Figure 4.24. Comparison of elongation at break (%) of original PVB and recycled PVB in blend with PVC, clay and calcium carbonate.

The selected polymeric films were examined by SEM analysis to evaluate surface morphology from the ruptured areas. Effects of different concentrations of PVB, PVC, clay, and $CaCO_3$ on surface morphologies of polymeric films after the mechanical test are given in Figure 4.25.

It can be seen from Figure 4.25 that mechanical surface morphology of original and recycled PVB polymeric films are very uniform and homogeneous. The mechanical surface morphology shows two separate interfaces and heterogeneous structure with the addition of 10 wt% of PVC into the original PVB. When 0.1 wt% of clay is added into OB90_P10, well-dispersed surface morphology is obtained. The polymeric film with the same amount of RPVB shows a rougher behavior than the blend with original PVB as well. The addition of 5 wt% calcium carbonate affects the surface morphology negatively than clay addition.

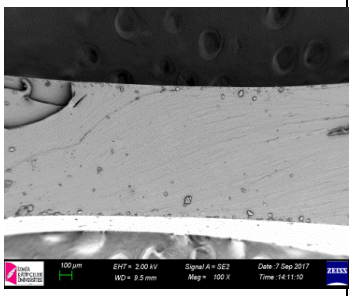
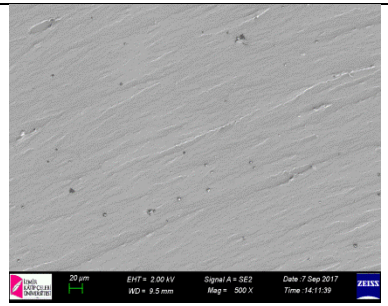
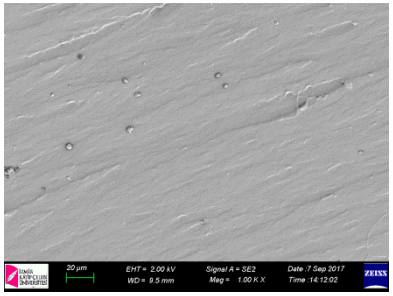
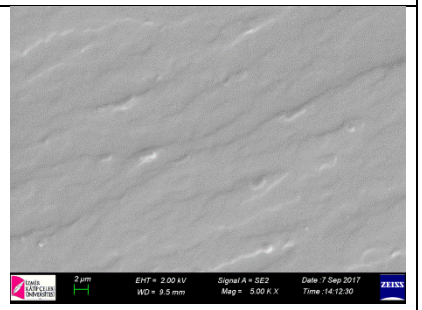
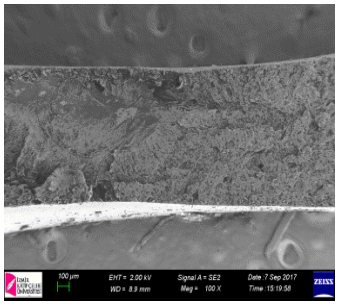
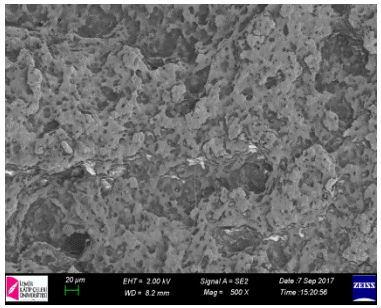
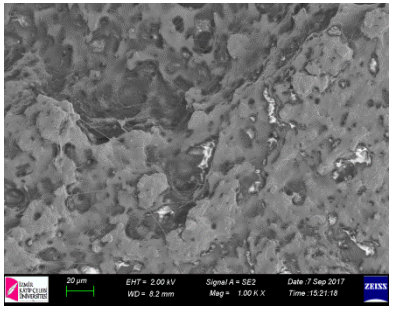
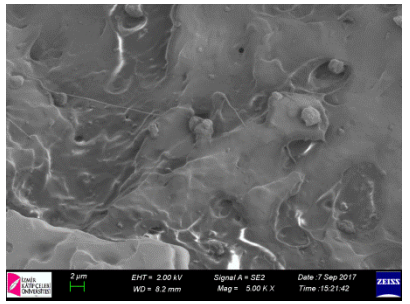
Film	100x	500x	1000x	5000x
OB100				
OB90_P10				

Figure 4.25. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on mechanical surface morphologies of polymeric films by SEM analysis.

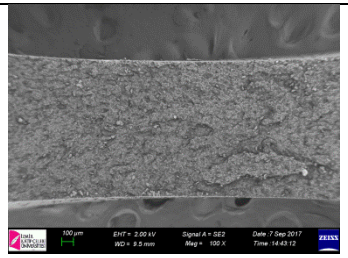
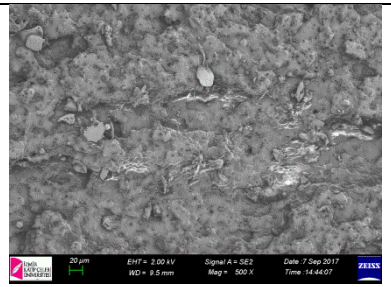
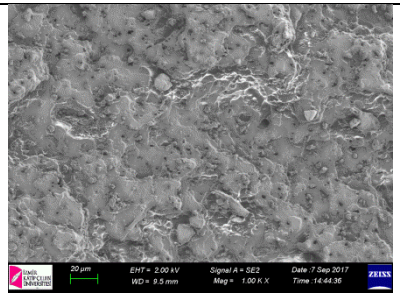
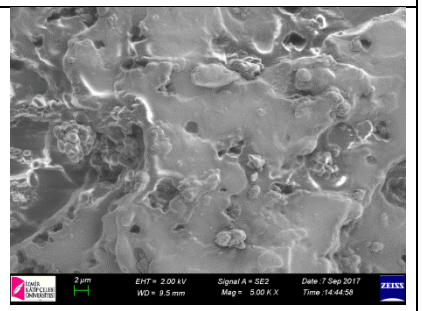
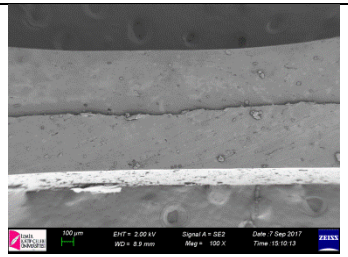
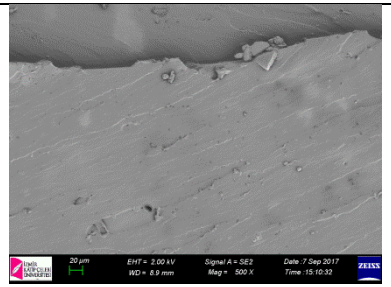
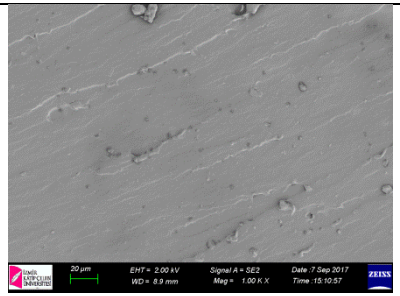
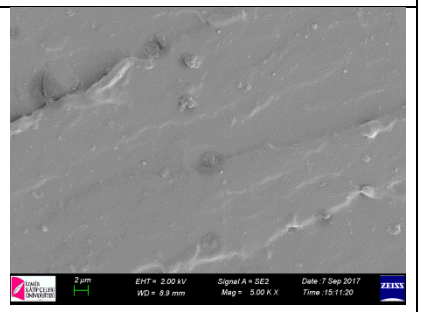
Film	100x	500x	1000x	5000x
OB90_P10_C0.1				
RB100				

Figure 4.25. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on mechanical surface morphologies of polymeric films by SEM analysis (continued).

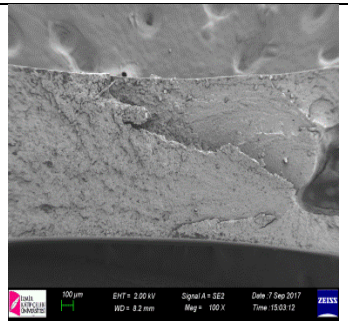
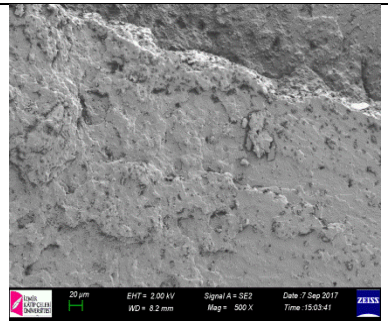
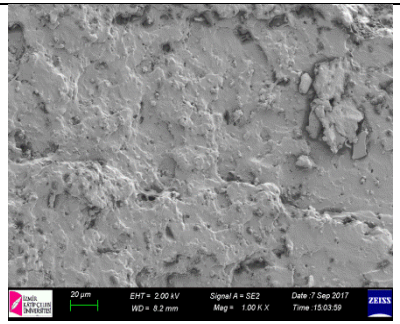
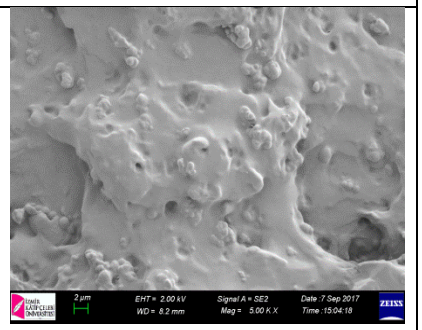
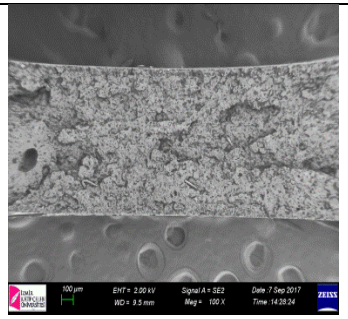
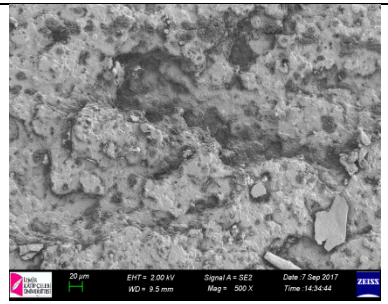
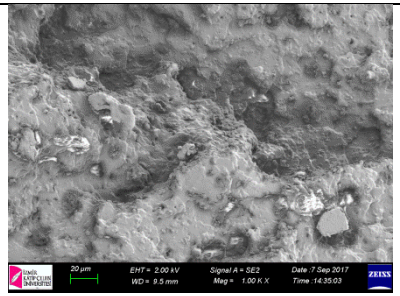
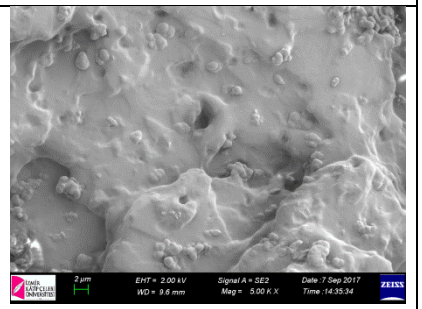
Film	100x	500x	1000x	5000x
RB90_P10				
RB100_P10_C0.1				

Figure 4.25. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on mechanical surface morphologies of polymeric films by SEM analysis (continued).

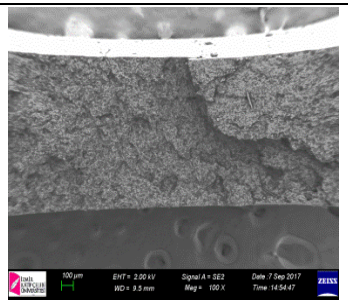
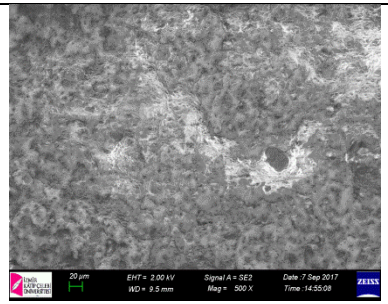
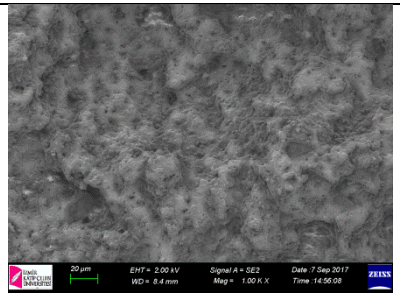
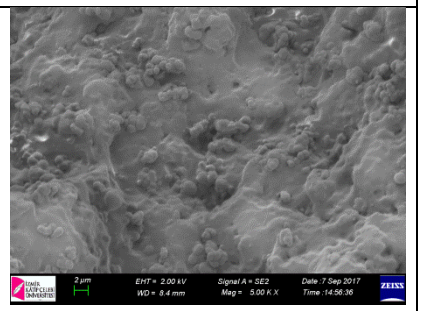
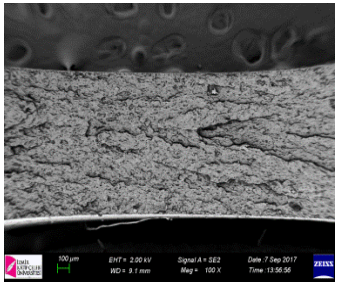
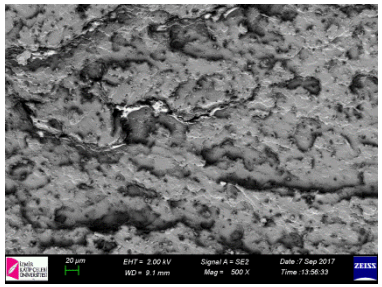
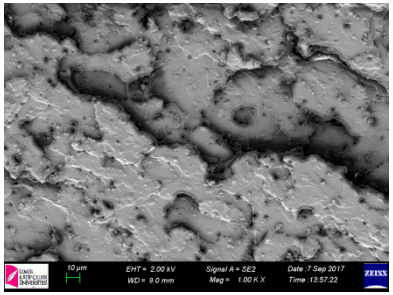
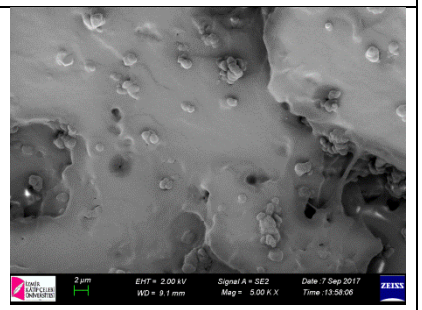
Film	100x	500x	1000x	5000x
OB80_P20_K5				
P100				

Figure 4.25. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on mechanical surface morphologies of polymeric films by SEM analysis (continued).

4.4 Thermal Properties

Figure 4.26 shows how selected polymeric films changed their color GXULQJ WKHUPDO GHJUDGDWLRQ EHWZHHQ DQG PLQXW
that thermal degradation of all polymeric films starts at 20 minutes. The polymeric films with recycled PVB are darker than with original PVB polymeric. Because recycled PVB was treated for the second time. The visual color of PVC did not change during thermal degradation but the changes in the physical structure were observed. The addition of 5 wt% of $CaCO_3$ into polymeric films caused very different color behavior (like a dried rose-pink color) during thermal degradation than others.

Figure 4.27 shows L*, a*, and b* values for polymeric films after thermal degradation. Lightness (L*) is generally decreasing through 60 minutes for all polymeric films. In the same time, the value of a* red color is increasing. There is also increment for b* value which represents yellow color.

It can be seen in Figure C1 in Appendix, at the beginning (0 min), the effect of the addition of 0.1 wt% and 1 wt% of clay on the color is not obvious, but for the addition of 5 wt% clay, the color yellow increases. Polymeric films with 1 wt% and 5 wt% clay added were almost black after 60 minutes. These results only show color changes during thermal degradation process.

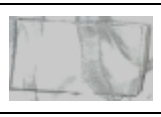
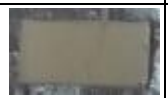
Sample Name	0 min	5 min	10 min	20 min	30 min	40 min	50 min	60 min
OB100								
OB90_P10								
OB90_P10_C0.1								
RB100								
RB90_P10								
RB90_P10_C0.1								
OB80_P20_K5								
P100								

Figure 4.26. Color changes of selected polymeric films after thermal degradation.

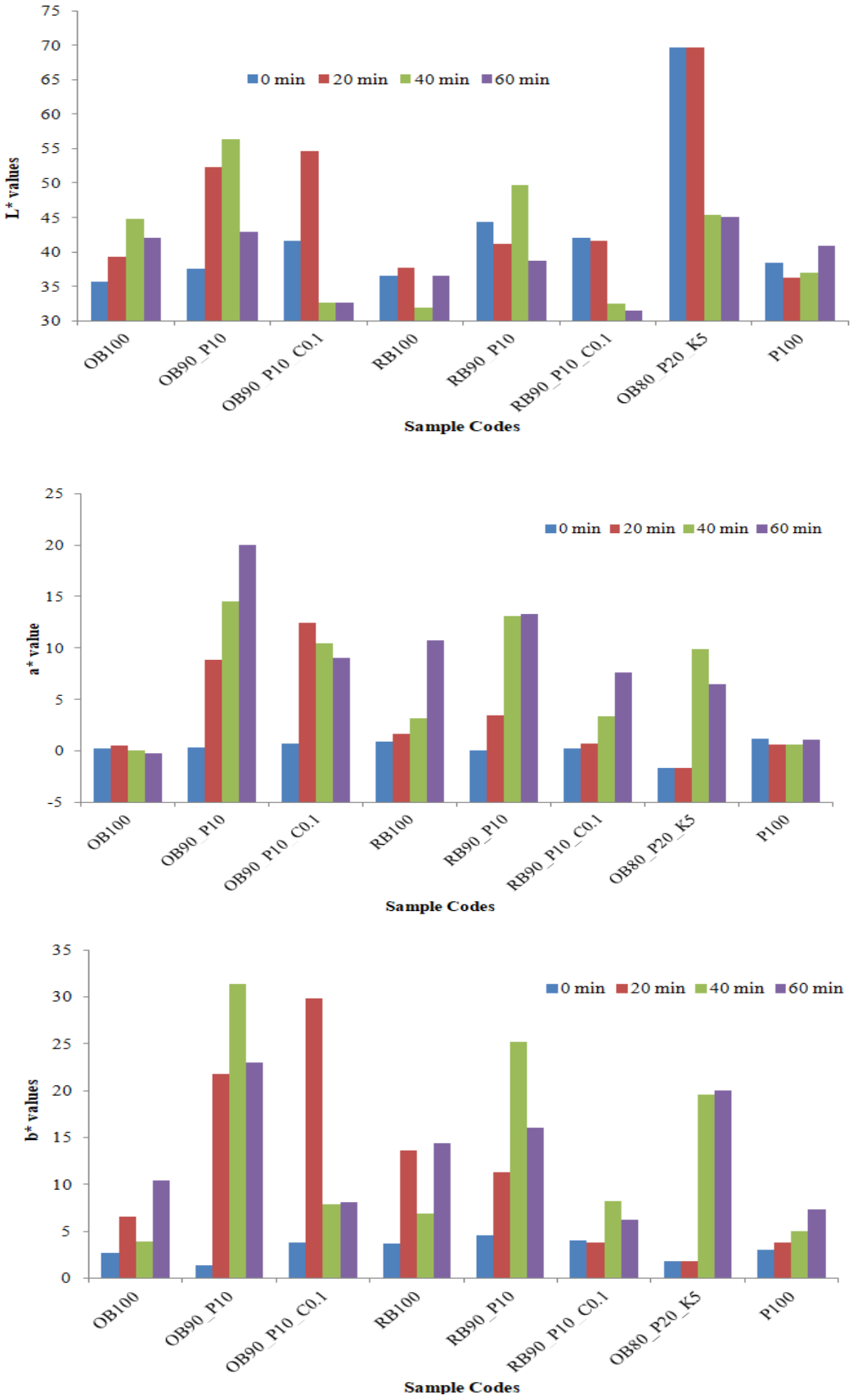


Figure 4.27. Comparison of L*, a* and b* values for the selected samples.

Chemical structure of polymeric film during thermal degradation was also evaluated. Figure 4.28 shows that FT-IR spectra of thermally degraded OB100 at f & I R 20-40-60 minutes and functional groups G L G Q f W during thermal degradation process.

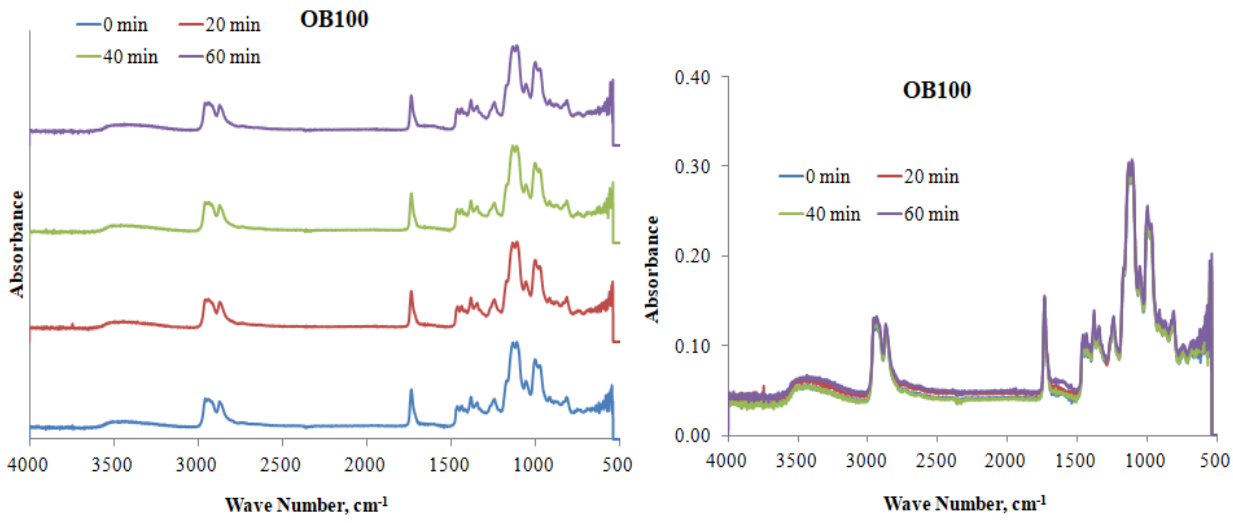


Figure 4.28. FT-IR spectra of W K H U P D O O \ G H J U D G H G 20-40-60 minutes. f & I R U

From Figure 4.29, it can be seen that functional groups of Recycled PVB stayed the same during thermal degradation process.

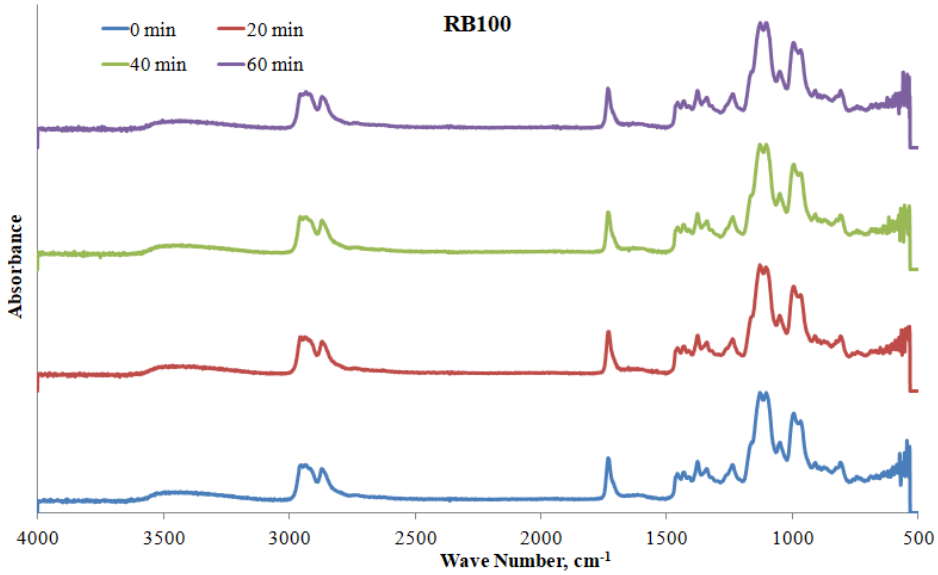


Figure 4.29. FT- , 5 V S H F W U D R I W K H U P D O O \ G H J U D G H G 5 % 20-40-60 minutes

Figure 4.30 shows that FT-IR spectra of plasticized PVC and there are some deformation because of HCl formation.

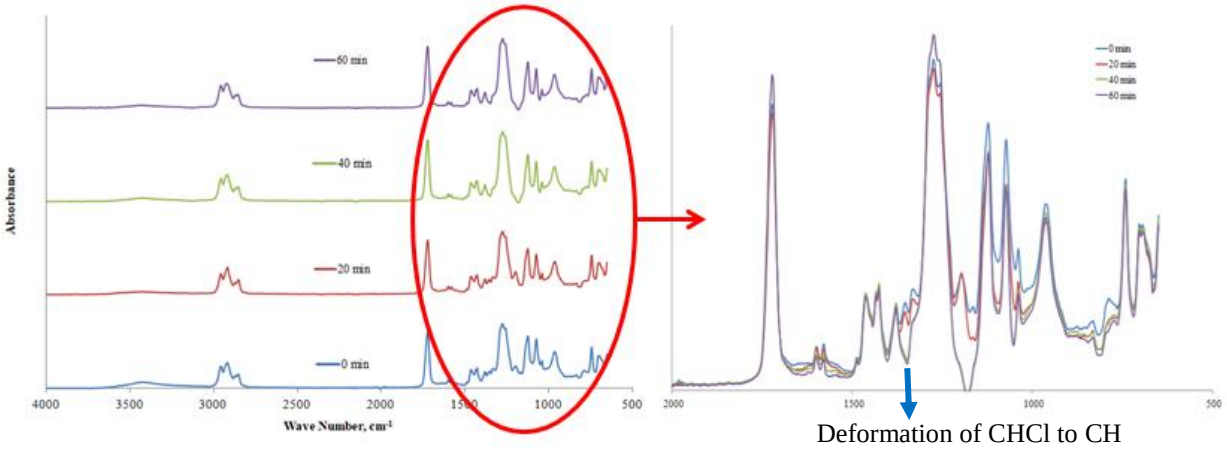


Figure 4.30. FT-IR spectra of thermally degraded PVC film at 0-20-40-60 minutes.

For OB90_P10, FT-IR spectra show slight deformation on the functional groups during thermal degradation process in Figure 4.31.

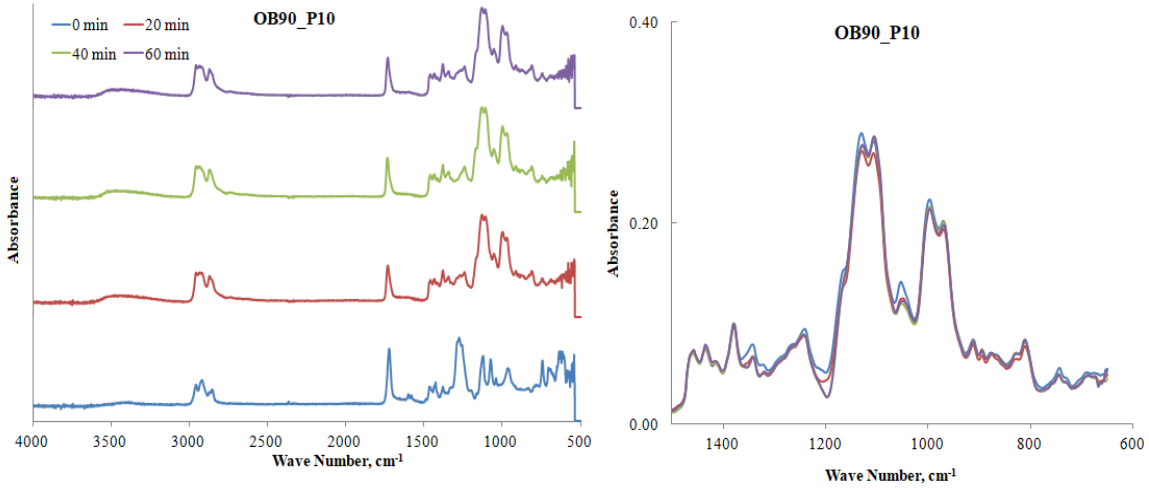


Figure 4.31. FT-IR spectra of thermally degraded OB90_P10 film at 0-20-40-60 minutes.

It can be seen from Figure 4.32 that the functional groups did not change during the thermal degradation process for OB95_C5.

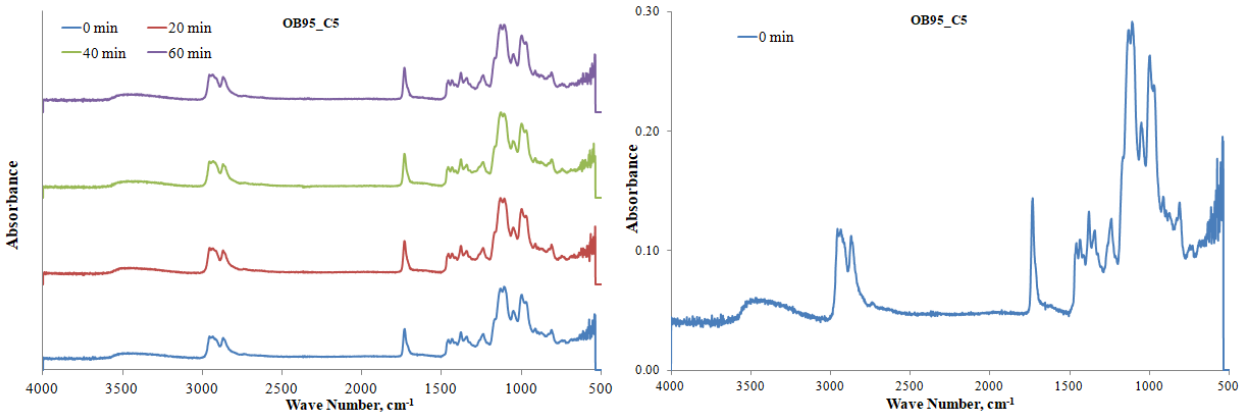


Figure 4.32. FTIR analysis profiles of thermal degradation for OB95_C5 at 0-20-40- P L Q X W H V. D W

Functional groups observed were the same during the thermal degradation process of OB90_P10_C0.1 in Figure 4.33.

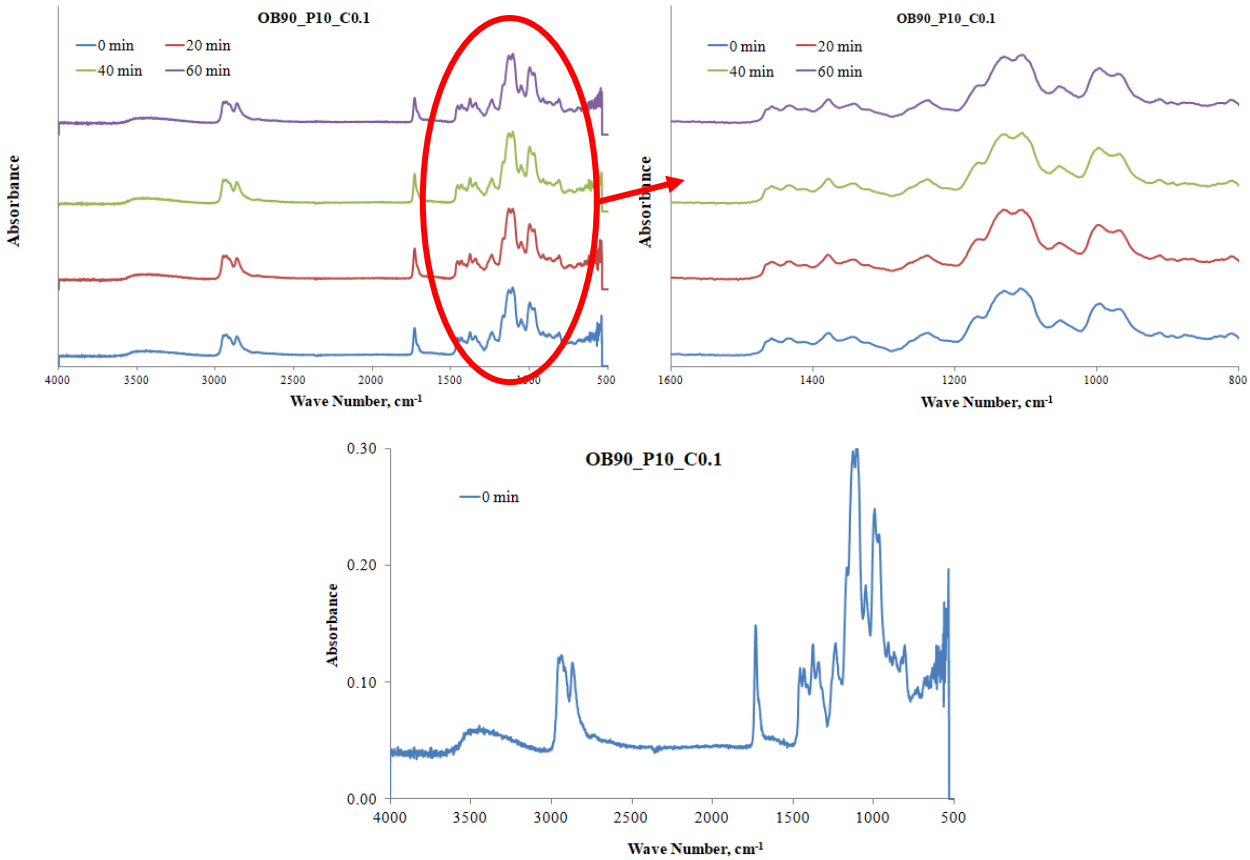


Figure 4.33. FTIR analysis profiles of thermal degradation for OB90_P10_C0.1 at 0-20-40-60 minutes at 170 ƒ &

XRD analysis of selected polymeric films was also evaluated during thermal degradation to observe the degree of crystallinity changes. As mentioned before, it was observed from the visual changes, the crystal structure and the degree of crystallinity did not change for OPVB at 60 min of the XRD pattern in Figure 4.34. This is the same also for RPVB as well. Both OPVB and RPVB are stable because of the good crystal structure and degree of crystallinity during thermal degradation. When the XRD pattern of OPVB and RPVB are compared after 60 minutes, it is not much different. It can be seen that PVC is much faster deteriorated than PVB at 60 minutes and is a very sharp degradation because of evaluation of hydro chlorination (HCl). Addition of 10 wt% of PVC into PVB has slightly decreased the degree of crystallinity and there are small differences between 0 and 60 minutes after thermal degradation.

The small amount of (0.1 wt%) clay improves the mechanical properties, but the degree of crystallinity is slightly decreased at 60 minutes from the XRD pattern. The same composition with recycled PVB has a lower degree of crystallinity than the original PVB at 60 minutes. Because recycled PVB was heat-treated for the third time. The degree of crystallinity is slightly decreased with the addition of calcium carbonate.

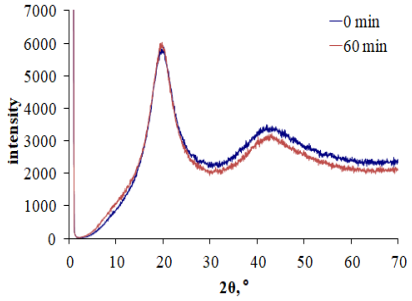
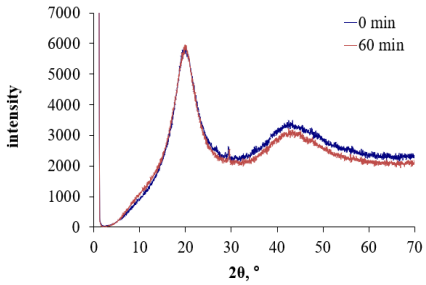
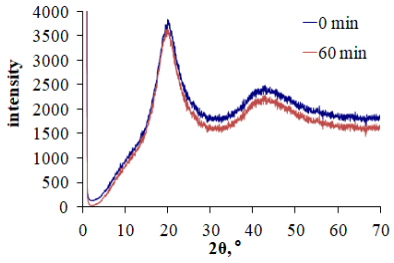
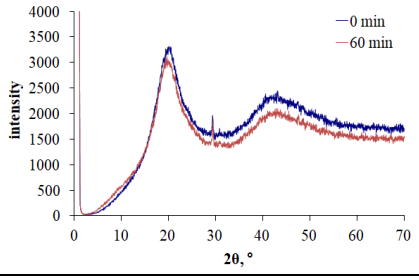
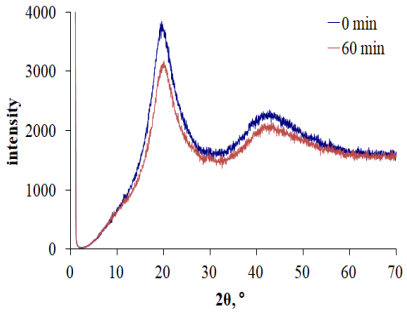
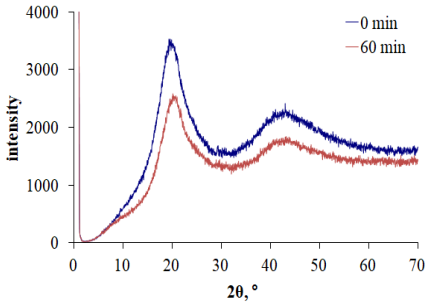
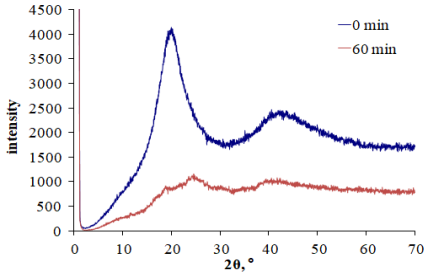
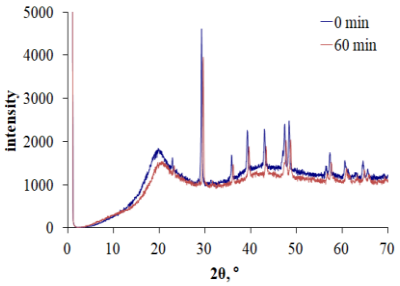
Sample Name	Original PVB	Recycled PVB
B100		
B90_P10		
B90_P10_C0.1		
P100		**not available**
OB80_P20_K5		**not available**

Figure 4.34. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition at 0 minutes and 60 minutes during thermal degradation.

Surface morphology of the original/recycled PVB and PVC with and without nano-additives films during the thermal degradation process was observed by optical microscopy and results are given in Figure 4.35 for the selected polymeric films.

It can be said that the thermal degradation of all polymeric films starts at 20 minutes. The original PVB is almost the same from 20 minutes to 60 minutes, and it has some crystallites after 20 minutes but as mentioned before the degree of crystallinity does not change after 60 minutes. Recycled PVB is dark at 40 minutes, but the degree of crystallinity does not change during thermal degradation. The changes in the structure were observed during thermal degradation for PVC. Addition of 10 wt% of PVC into original and recycled PVB caused color changing at 40 minutes. Surface morphology gives better homogenous and smooth results with the addition of 0.1 wt% of clay than without clay addition during thermal degradation behavior. The addition of calcium carbonate has caused black color and some deformations on the surface morphology than others.

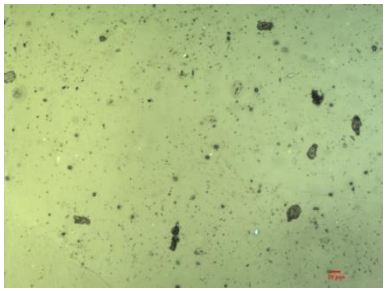
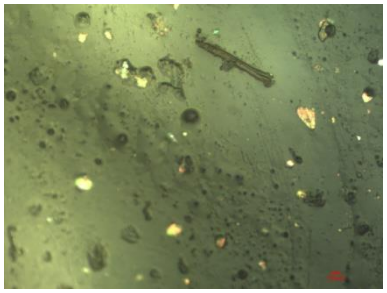
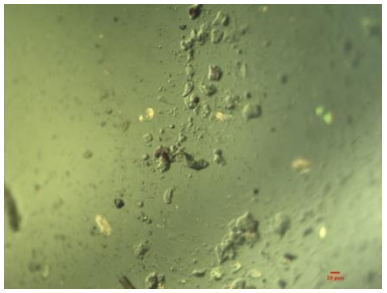
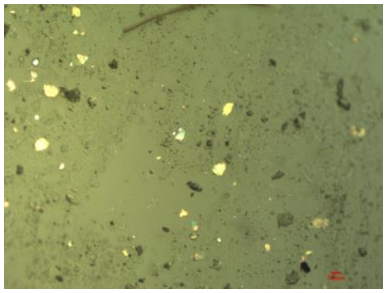
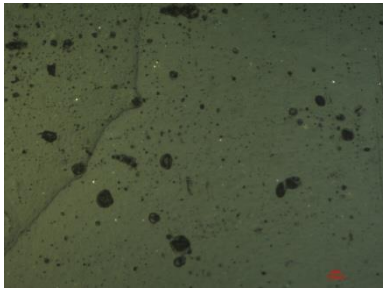
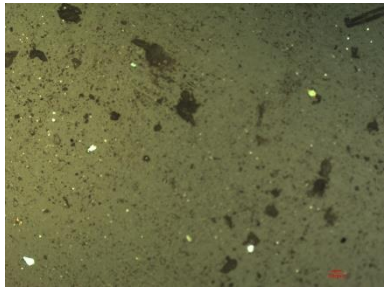
Sample Name	0 min	20 min	40 min	60 min
OB100				
OB90_P10				

Figure 4.35. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on surface morphologies of selected polymeric films during the thermal degradation by Optical Microscopy.

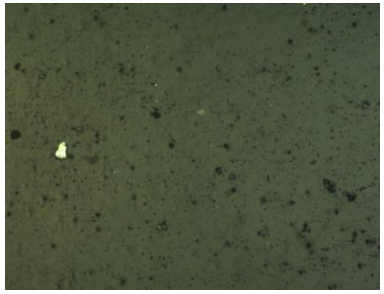
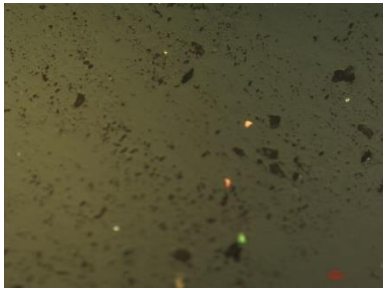
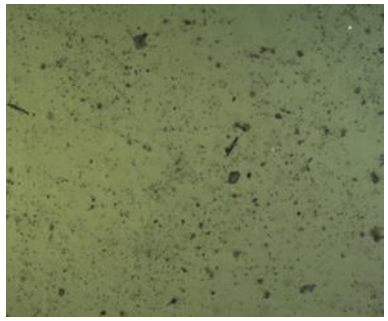
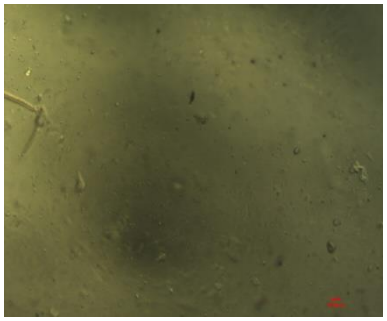
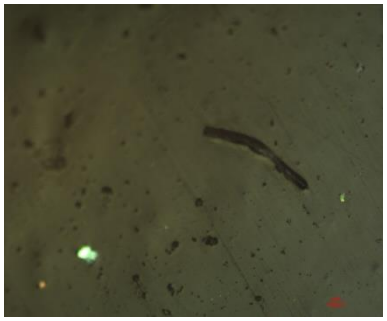
Sample Name	0 min	20 min	40 min	60 min
OB90_P10_C0.1				**not available**
RB100				**not available**

Figure 4.35. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on surface morphologies of selected polymeric films during the thermal degradation by Optical Microscopy (continued).

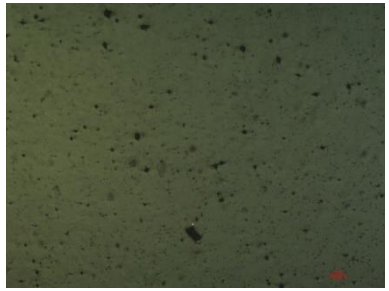
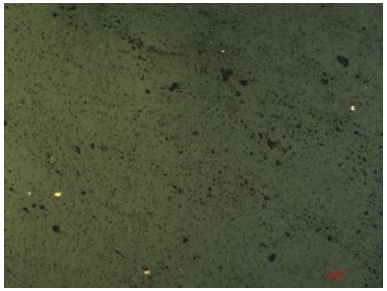
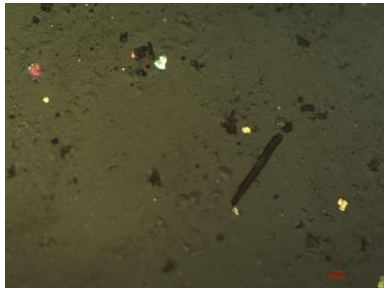
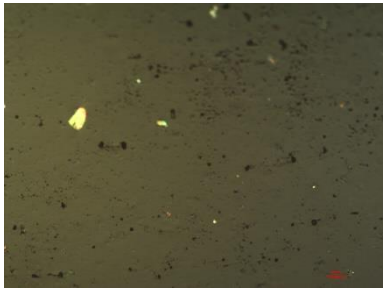
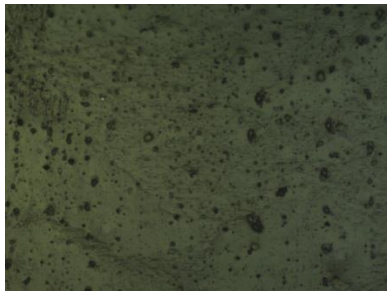
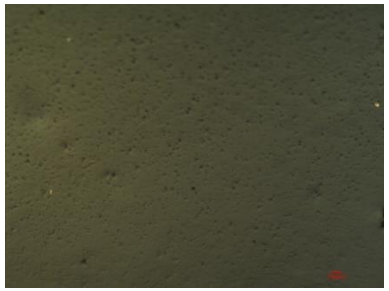
Sample Name	0 min	20 min	40 min	60 min
RB90_P10				
RB90_P10_C0.1				**not available**

Figure 4.35. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on surface morphologies of selected polymeric films during the thermal degradation by Optical Microscopy (continued).

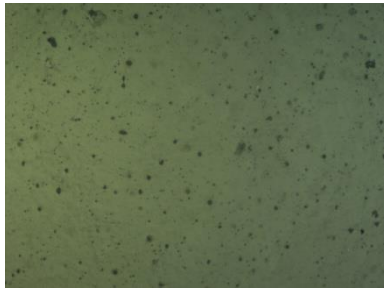
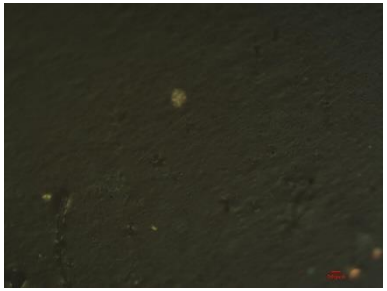
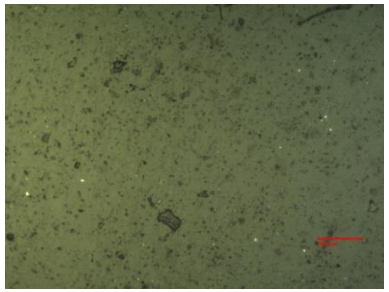
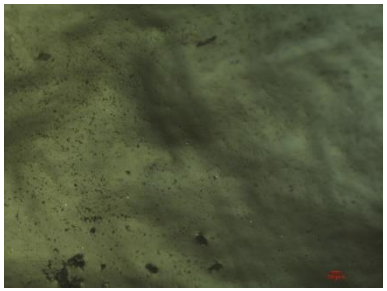
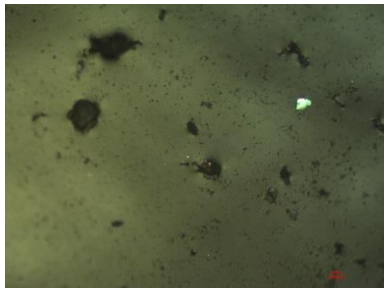
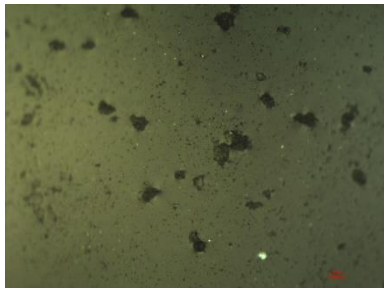
Sample Name	0 min	20 min	40 min	60 min
OB80_P20_K5				**not available**
P100				

Figure 4.35. Effects of different concentrations of PVB, PVC, clay and $CaCO_3$ on surface morphologies of selected polymeric films during the thermal degradation by Optical Microscopy (continued).

¾ **TGA Analysis**

Thermal degradation behavior of polymeric films is also analyzed by TGA thermograms which are given in Figure 4.36. The thermal decomposition was carried out under nitrogen atmosphere from 30 ƒ & WR ƒ & DW D KHDWLQJ UDW ƒ & PLQ EXW 39% completely thermally decomposed (in nitrogen) at 500 ƒ &

From Figure 4.36, weight loss of PVB occurs in two distinct regions between 200 ± 300 and 300 ± ƒ & DQG FRUUHVSRQGLQJ WR DERXW loss. The PVB was primarily losing plasticizer in the temperature volatilization process between 200- ƒ & the major products of the decomposition above 300 ƒ & ZHUH LQLWLDOO\ EXW\UDOGHK\GH DQG EXWHQDO EXW\UDO JURXSV IROORZHG E\ DFHWLF DFLG DERYH acetate groups. The thermal degradation of PVC occurs through two degradation steps. The temperatures at the maximum weight losses of PVC were observed at 330 and 500 ƒ & 7KH ILUVW VWHS FRUUHVSRQGV WR WKH dehydrochlorination of PVC, while the second step presents the total weight loss resulted from the degradation of the dehydrochlorinated residuals. Onset WHPSHUDWXUH RI SRO\PHULF ILOPV ZKLFK DUH 2% 2% B3 ƒ & 5% B3 ƒ & 2% ƒ & B3 B& 2% B3 B& ƒ & 2% B3 B. ƒ & ZHUH GHWHU KLJKHU RQVHW WHPSHUDWXUH , therefore with thermal stability is the best.

7KH DPRXQWV RI DVK YDOXHV DW ƒ & DUH D clearly understand the results. The amounts of ash % of original PVB (OB100) and recycled PVB (RB100) were found as 1.30 % and 3.16, respectively at 600 ƒ & was seen that the original PVB has a smaller value than others with respect to the amount of ash % value. Remaining weight % of PVC (P100) was found as DW ƒ & 7KH UHVXOWV RI DVK YDOXHV DUH with addition of PVC into original and recycled PVB but, it is lower than PVC. The addition of 0.1 wt% of clay is slightly decreased the amount of ash % value than original and recycled B90_P10 polymeric films. The result of ash content by the addition of calcium carbonate is higher than other polymeric films and the best flammability is obtained.

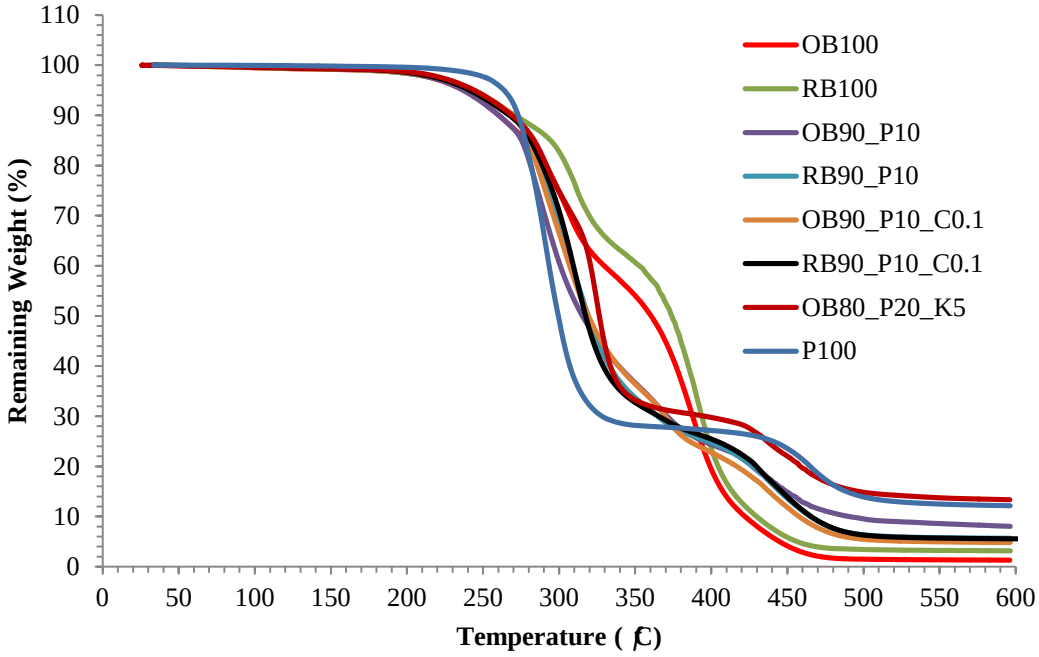


Figure 4.36. Thermograms of selected polymeric polymeric films.

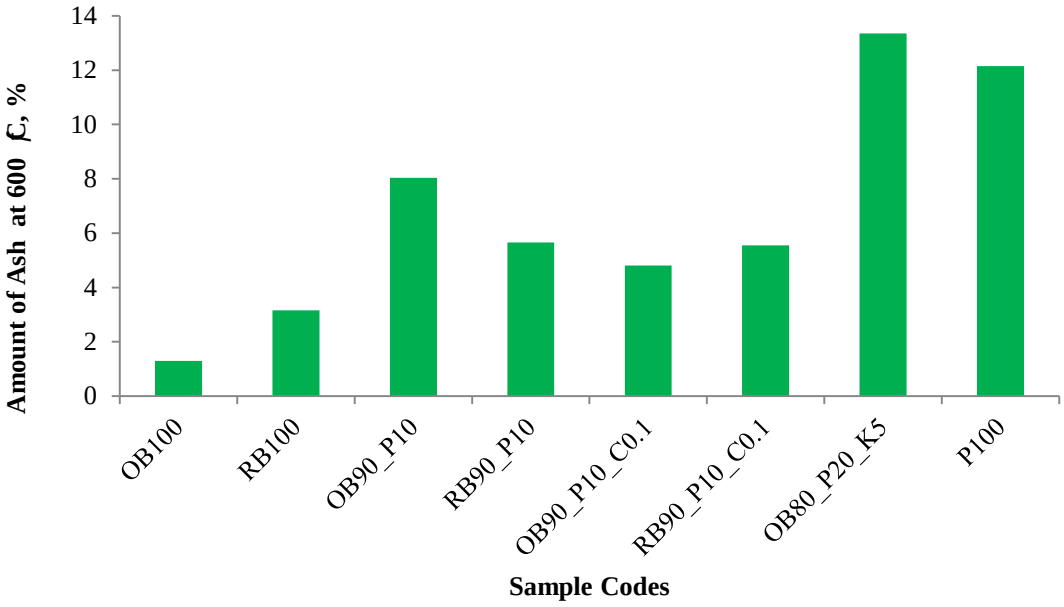


Figure 4.37. 7KH DPRXQW RI DVK DW ƒ&

¾ **DSC Analysis**

The DSC analysis determined that a glass transition temperature (Tg) of RULJLQDO 39% ZDV DOPRVW ƒ& DQG OLWWOH \FOHG 39 higher as shown in Table 4.7. The glass transition temperature of PVC was around - ƒ& ZKLFK ZDV much lower glass transition temperature than PVB. The glass transition temperature of the addition of PVC into PVB polymeric film is actually expected that it should lower the value, but it was found as increased thanks to beginning good molecular interaction. However, obtaining a single glass transition temperature shows that the films have high compatibility.

Table 4.7. Results of DSC analysis for original and recycled PVB/PVC polymeric films

	Original PVB	**Recycled PVB**
Sample Name	**Glass** 7UDQVLW	*ODVV 7UDQV
B100	17.77	22.75
B80_P20	18.06	38.44
B50_P50	26.76	28.93
B20_P80	34.19	25.21
P100	-25.69	-25.69

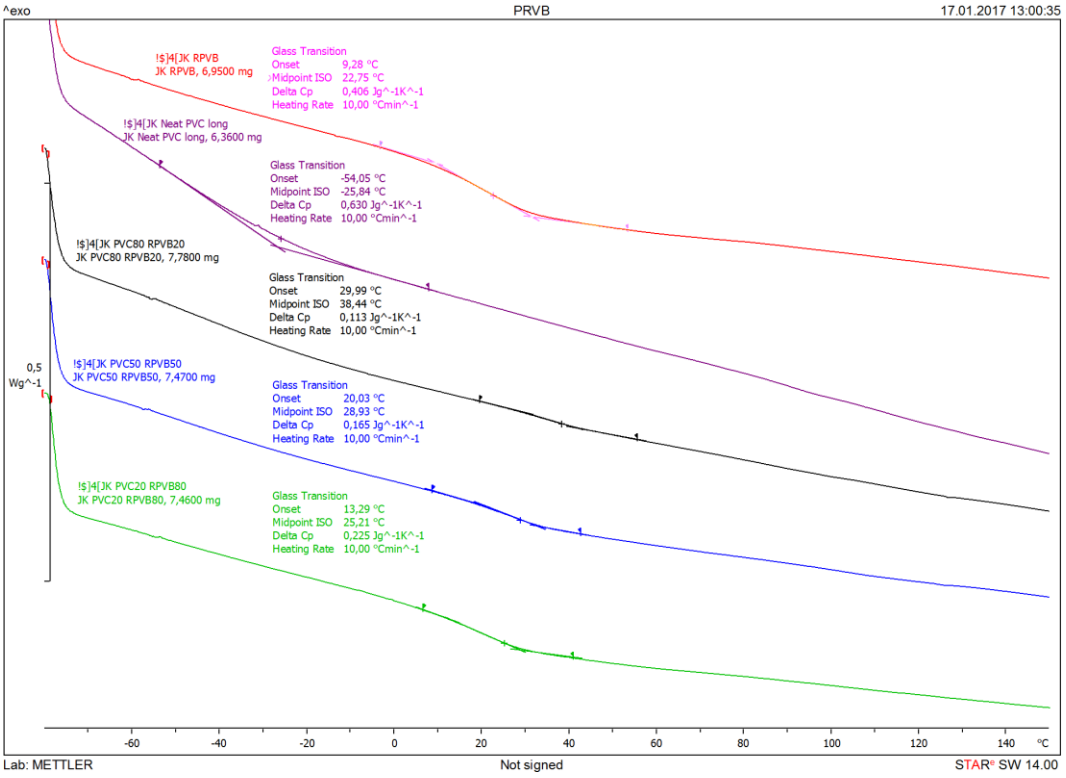


Figure 4.38. DSC profile of selected polymeric composite film with recycled PVB.

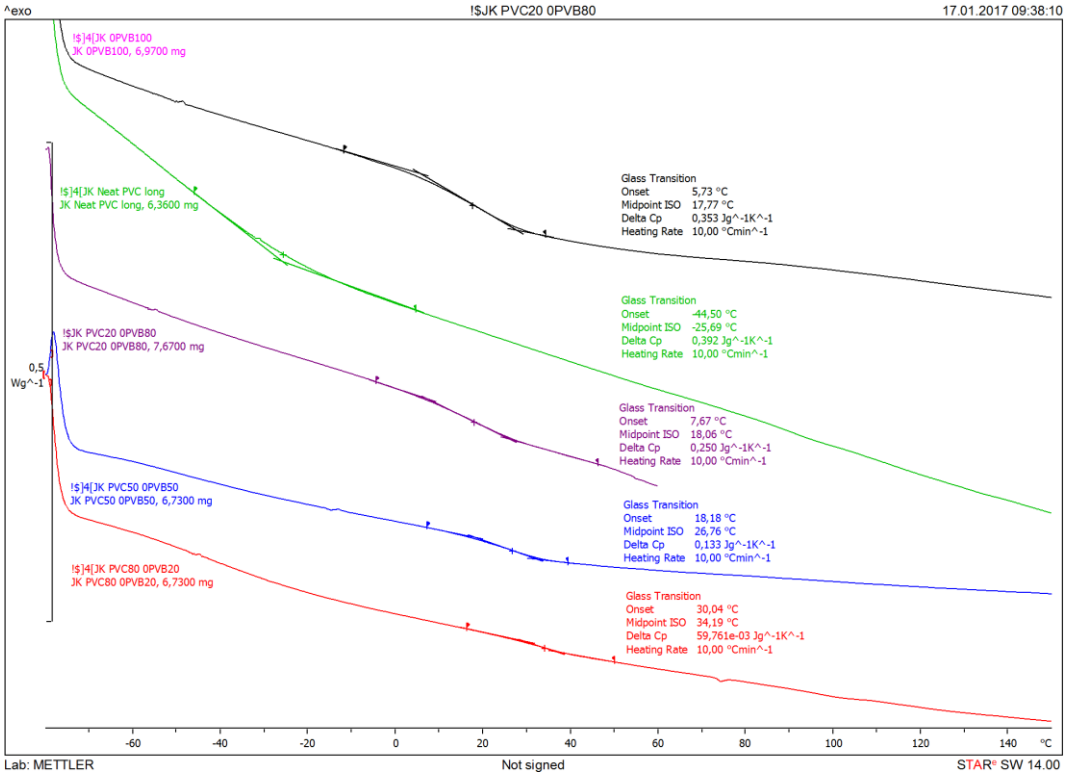


Figure 4.39. DSC profile of selected polymeric composite film with original PVB.

5. CONCLUSION

In conclusion, investigating the possibility of new applications of recycled PVB will help to solve a major environmental problem and decrease the price of traditional PVB and PVC as obtained good product properties of polymeric films that can be seen below:

- The inter glassed PVB sheets possess physical properties very similar to those of the extruded material before the lamination process.
- It can be mentioned that as 0.1% (wt/wt) Clay added OB90_P10 and RB90_P10 polymeric films showed the highest tensile strength and highest elongation at break when compared to the other additive and amounts based polymeric films , W ¶ V S URB90_P10_C0.1 film has good tensile strength than commercially used PVC materials.
- FT-IR spectra of original plasticized PVB obtained from literature and in this study are really consistent. The functional groups of the original and recycled films are the same.
- The FT-IR profiles are very similar to each other. The polymeric films are uniform and no formation of a new functional group is observed. It means during the production the chemistry of the polymers did not change.
- Addition of 0.1% clay has decreased the degree of crystallinity a little bit but it has given very good mechanical properties.
- 5 wt% amount of clay addition has decreased the degree of crystallinity.
- Additions of 5 wt% $CaCO_3$ into polymeric films have given better mechanical results than addition of 5 wt% clay.
- The polymeric film color of PVC was unchanged, but its crystallinity was reduced after the thermal degradation process.
- Surface morphology of recycled PVB shows more heterogeneous structure than the original PVB because recycled PVB has various impurities.
- The surface morphology is a bit heterogeneous with the addition of 10 wt% PVC into PVB. The addition of 0.1 wt% clay has similar behavior at surface morphology.

- It can also be said that surface morphology of the all obtained polymeric films with recycled PVB show more heterogeneous structure than the original PVB.
- PVC has a higher T_{onset}, therefore its thermal stability is the best.
- The result of ash content by the addition of calcium carbonate is higher than other polymeric films and the best flammability is obtained.
- Obtaining a single glass transition temperature shows that the films have high compatibility.
- Consequently, recycled PVB/PVC/Clay based composite films can be recommended as a new material instead of PVC.

ACKNOWLEDGEMENT

I express my deep gratitude to my supervisor Prof. Dr. Serap CESUR for their valuable advices, times, guidance and patience in the completion of this thesis.

I would like to give my special thankfulness to the financial support provided by Erasmus + Program of EU to avail the facilities of Tomas Bata University in Zlin, Czech Republic. This program has provided me different background and perfect academic skills. For this reason, I am grateful to My Erasmus+ supervisor Assoc. Prof. Dr. Dagmar 0 tQ Vwi has shared her valuable academic background and ' U $ O L F H 7-6 V E ERIGRY i her kind help in the experimental and characterization work.

I would like to indicate very special thanks to Chemical Engineering Department, Ege University for allowing me to use the test laboratories. I offer our special thanks to chemist Korcan Korba for his help in FTIR analysis.

Finally, I am faithfully grateful to my family and my friends for continuous support and encouragement.

31/07/2019

&DQVX .g52ö/8

REFERENCES

Axinte, E., 2011, Glasses as engineering materials: A review, Materials&Design, 32(4), 1717-1732pp.

Bendaoud, A., Carrot, Ch., Charbonnier, J., Pillon, C. ³ %OHQGV RI Plasticized Polyvinyl Butyral and Polyvinyl Chloride: Morphology Analysis in View of Recycling, Macromol Symp Eng, vol. 298, pp. 1259-1268pp.

Carrot C., Bendaoud A., Pillon C., 2015, Polyvinyl Butyral from: Handbook of Thermoplastics CRC Press, 89-137pp.

Castori G., Speranzini, **E.**, 2017, Structural analysis of failure behavior of laminated glass, Composites Part B 125, 89-99pp.

Cheremisinoff, P. N., 1998, Advanced Polymer Processing Operations, Noyes Publications, Wesiwood, New Jersey, U.S.A., 40p.

Dhaliwal, A.K., Hay, J.N. ³ 7KH characterization of poly(vinyl butyral) by WKHUPDO DQDO\VLV ¶¶ -RXU7KHUPDO$QDO&DO YRO 25pp.

Fernandez, M. D., Fernandez, M. J., **Hoces, P.** ³6\QWKHVLV RI 3RO\ YLQ\O EXW\UDO V LQ +RPRJHQHRXV 3KDVH DQG 7KHLU 7KHUPL Polym. Sci. 102, 5007-5017pp.

Herrmann, W. O., Haehnel, W., 1927, Uber den Poly-vinylalkohol. Ber Deut Chem Ges (A and B Ser) 60, 1658 ±1663pp.

IOCA, The International Organization of Motor Vehicle Manufacturers, http://www.oica.net/category/production-statistics/2018-statistics/ (Date of Access: 18.01.2019)

Lindeman, M. K., 1989, Vinyl acetate and the textile industry. Text Chem Color 21, 21 ±28pp.

REFERENCES (continued)

Mohammadian-Kohola, M., Asgari H.R., Shakur, H.R., 2016, A detailed investigation of the gamma-ray radiation effects on the optical properties RI SRO\YLQ\O EXW\UDO=746pp 2SWLN

Nascimento, M. L. F., 2014, Brief history of the flat glass patent sixty years of the float process, World Patent Information 38 50-56pp

Nascimento, M. L. F., Zanotto, E. D., 2016, On the first patents, key inventions and research manuscripts about glass science & technology, World Patent Information 47, 54-66pp.

3DLYD / % 0RUDOHV $, 2008, Organoclays: Properties, preparation and applications, Applied Clay Science, 42, 8-24pp. DQG 'D G) 5

Patrick, S. G., 2005, Practical Guide of Polyvinyl Chloride, Rapra Technology Limited, Shropshire, United Kingdom, 17p.

Patterson, M., 2011, Structural Glass Facades and Enclosures. John Wiley & Sons, Inc., Hoboken, New Jersey.

Paul, D., 1989, Control of phase structure in polymer blends, in Functional Polymers (eds D.E. Bergbreiter and C.R. Martin). Plenum Press, s. 1-18pp.

Rhim J. W., and Ng P. K. W., 2007, Natural Biopolymer-Based Nanocomposite Films for Packaging Applications, Critical Reviews in Food Science and Nutrition, 47, 411-433pp.

6DYLü - - 0LXMRLYüL ü ' % R J 2013, Architectural Glass: Types, Performance and Legislation, Architecture and Civil Engineering Vol. 11, No 1, 35 - 45pp.

REFERENCES (continued)

Schnell, R. J., Ayala, D. S., Meerow A. W., Winterstein M., Cervantes C., Brown, J. S., 2005, Determination of color and fruit traits of half-sib families of mango (Magnifera indica L.). Proc. Fla. State Hort. Soc., 118 253-257pp.

6HUDILQDYLþLXVD 7 /HEHW - 3 /RXWH,U & /HQNLPDV 2010, Long-term laminated glass four point bending test with PVB, EVA and SG interlayers at different temperatures, Procedia Engineering 57, 996 ±1004.

Tupy, M., Merinska, D., Svoboda, P., Zvonicek, J., ³,QIOXHQFH RI ZDWHU and magnesium ion on the optical properties in various plasticized SRO\ YLQ\O EXW\UDO VKHHWV´ -RXU$SSO3RO\P6FL YR

Tupy, M., Merinska, D. and Kasparkova, V., 2012. PVB sheet recycling and degradation. In Material Recycling ² Trends and Perspectives, Chap. 5, ed. D. Achilias, Rijeka, Czech Republic: InTech.,133 ±150pp.

Tupy, M., Mokrejs, P., Merinska, D., Svoboda, P., Zvonicek, J., 2014a, Windshield recycling focused on effective separation of PVB sheet, JourApplPolymSci, vol. 131, issue 4.

7XS\ 0 0HULQVNDG6YRERGD3 &DUURW & 3LOORQ 3HWUiQHN 9E ³0HFKDQLFDO 3URSHUWLHV RI 5HF\FOHG 39% 39& %OHQGV´ :RUOG $FDGHP\ RI 6FLHQFH (QJ Technology, International Journal of Civil, Environmental, Structural, Construction and Architectural Engineering Vol:8, No:9.

Wade, B., 2004, Vinyl acetal polymers. In Encyclopedia of Polymer Science and Technology, 3rd Ed., Vol. 8, ed. H. F. Mark, 381 ±399. New York: John Wiley & Sons.

Wade, B.E., D'errico, J.J., Thompson, H.M., Yu, M.K., 2004, Polymer sheets and methods of controlling adhesion of polymer sheets to glass, US Patent Application US 20050208315, Mar 16.

REFERENCES (continued)

Wang, H., Xie, C., Yu, W. and Fu, J., 2012, Efficient combined method of selective dissolution and evaporation for recycling waste polyvinyl butyral films. Plast Rubber Compos 41, 8 ±12pp.

: H O O H U % : • Q V F K ,-2005, Experimental Study on Different Interlayer Materials for Laminated Glass. EVA. Glass Processing Days, 124-127 pp.

Yu, J., Sun, L., Chuan Ma, Qiao, Y., Yao, H., 2016, Thermal degradation of PVC: A review, Waste Management 48, 300 ±314pp.

Zang, M. Y., Chen, S. H. / D P L Q D W H G * O D V
10.1002/9781118097298.weoc121

CURRICULUM VITAE

PERSONAL INFORMATION

Name-Surname : Cansu Göröl

Date of birth : 17.12.1991

Place of birth : Ordu

Nationality: : T.C.

Marital Status: : Single

Driver's License : Has (2013)

Adress : Ergene mah., 544.Sokak No: 21 K: 3 D: 7 Seyhan apart.
Bornova / İzmir

Phone Number : 0541 223 98 11

E-mail : cansukoroglu@hotmail.com / cansukorogluu@gmail.com

WORK EXPERIENCES

2017 : Bakanlıklar Denetim Elemanı (Quality Control Engineer) (06.11.2017 - «

2017 : UCTEA Chamber of Chemical Engineers Aegean Branch,
Symposium Secretary (12.05.2017 ±05.11.2017)

2014 : Ordu Yardımlaşma Kurumu, Summer Internship (1 month)
Sinop Boyalı Kumaş Sanayi Ltd. Şti. Summer Internship (1 month)
Mopak Kağıt Ve Ambalaj San. Ltd. Şti. Summer Internship (1 month)

2012 : Holiday Inn Cleveland-West (Westlake), Ohio ±ABD, Housekeeping (3 months)

2011 : Ahenk Sanat Merkezi, Secretary (4 month)

EDUCATION

2015-2019 Ege University, Chemical Engineering, MSc / English-3.43/4

2016-2017 Tomas Bata Univerzita in Zlín, Polymer Engineering Technology, Czech Republic (5 months). Master Thesis performed as Erasmus Student.

2009-2015 Ege Üniversitesi Chemical Engineering Bachelor

2005-2009 Değerli Lisesi / 98.96/100

1997-2005 Feyza Paşa İlköğretim Okulu

COMPUTER SKILLS

- Microsoft Office (Word, Excel, PowerPoint)
- Matlab
- Bricsnet IntelliCAD2000
- ChemCad
- Polymath

FOREIGN LANGUAGES

English :

- Ege University, School of Foreign Languages Preparatory School
- EGESEM English Speaking Club Certificate
- Writing, reading, speaking (intermediate)

PUBLICATIONS & PROJECTS

- &HVXU 6 .|UR÷<OXOoÕQ DQG Antimicrobial and Biodegradable Food Packaging Applications of Polycaprolactone/Organo nanoclay/Chitosan Polymeric Composite Films, Journal of Vinyl & Additive Technology, doi: 10.1002/vnl.21607, 376-387pp.
- Master Thesis "Original and waste Polyvinyl Butyral (PVB) / Polyvinyl Chloride (PVC) composites with nanoadditives and the degradation behaviour" which is carried out in Tomas Bata University of Department of Polymer Engineering, under the Erasmus+ co-advisor of Assoc. Prof. 'U 'DJPDU 0HUtQVsNi ILYH PRQWK
- Diploma Project "Antimicrobial and Biodegradable Food Packaging Applications of Polycaprolactone/Chitosan Polymeric Composite Films" which is supported by the Scientific and Technological Research Council RI 7XUNH\ 7h%ø7$. WKURX(2014-2015). SURMHFW

SYMPOSIUMS AND CONGRESSES

- 5th International Polymeric Composites Symposium and Workshops, ø]PL2-5 November 2017, Cansu .|UR÷Sexp Cesur, Dagmar 0 tQ,V Enhancement of Product Properties of Original and Waste

Polyvinyl Butyral (PVB) / Polyvinyl Chloride (PVC) Composites with Nano-Additives ´(orally presentation).

- ø1 2 9 $ 6 < 2+1 DIWDVÕ 17-18 March 2016, &DQVX .|UR÷OX (UJ•Q 'R÷UXNDú *L]HP ,úÕN 6HUDS &HVXU ³3ROLPHULF Composites in Antimicrobial and Biodegradable Food Packaging Film Applications, Project 1- The Production of Antimicrobial Food Packaging Films, LDPE/Chitosan/Nanoclay, Project 2- The Production of Biodegradable Food Packaging Films, /'3(3&/ 1DQRNLO.LWRVDQ (poster presentation).
- 32/ø.20 ,QWHUQDWLRQDO :RUNVKRS RQ 6SHFLDO 7RS Composites, February, 24-26, 2016, ø]PLU 7&DQVX .|UR÷OX %XUFX 6ÕUPD 6HUDS &HVXU ³.LWRVDQ g]HOLNOHUL YH 8\J (oral presentation).
- KOMPEGE III, 05- 1RYHPEHU -.XúDGDVÕ øzPLU "Antimicrobial and Biodegradable Food Packaging Applications of Polycaprolactone/Chitosan Polymeric Composite Films" (orally presentation).
- 7•UNL\H øQRYDV\RQ +D-Ewa DayÕ øzPLU December øVWDQEXO ³3RO\ODNWLNDVLG Kitosan Kompozit Filmlerinin Antimikrobiyal ve %L\RER]XQXU *ÕGD $PEDODMÕ 8\JXODPDODUÕ (poster presentation).
- .LP\HYL 0DGGHOHU YH 0DP•OHU 3URMH 3D]DUODPD *U $U øVWDQEXO 1RYHPEHU ³3OLNDSURODNWRQ .LWRVDQ Filmlerinin AQWLPLNUREL\DO YH %L\RER]XQXU *ÕGD $PEDODM (poster presentation).

INTERESTS AND HOBBIES

Playing Piano, acting and teaching in Theatre, Painting

REFERANCE

Prof. Dr. Serap Cesur
Ege University, Chemical Engineering Department
Phone Number: 0232 311 4045, 0533 462 4077
E-mail: serap.cesur@ege.edu.tr, cesur.serap@gmail.com

APPENDICES

APPENDIX A. Optical Microscopy

APPENDIX B. Mechanical test results

APPENDIX C. Thermal Degradation

APPENDIX D. X-Ray Diffraction (XRD) Analysis

A. Optical Microscopy

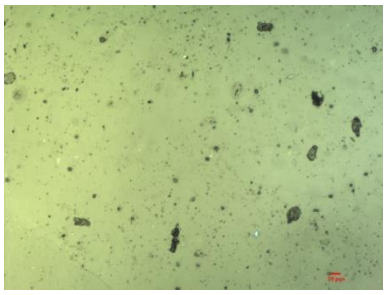
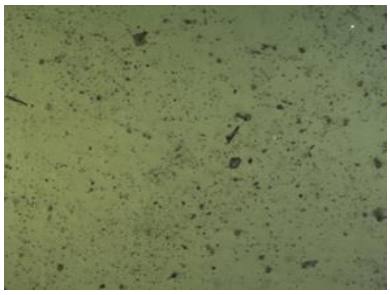
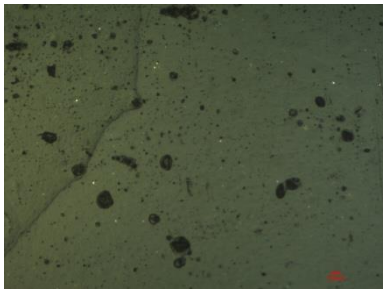
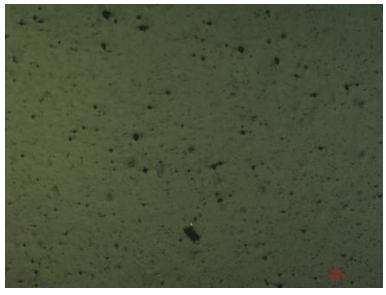
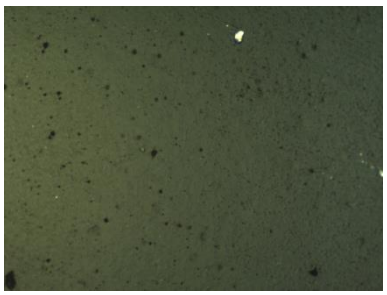
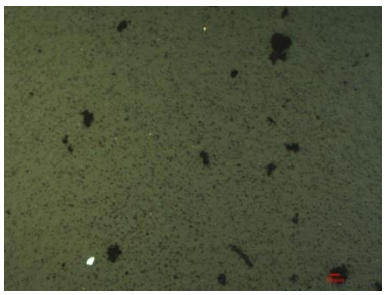
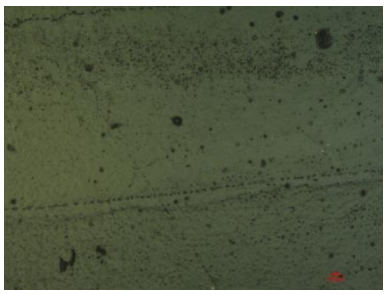
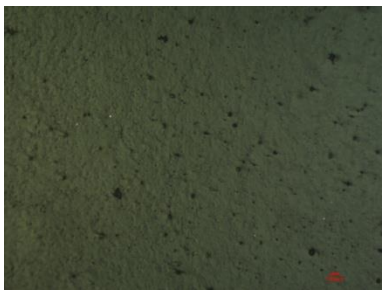
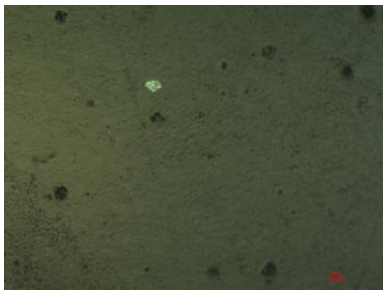
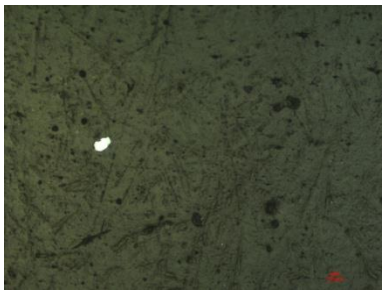
Sample Name	Original PVB	Recycled PVB
B100		
B90_P10		
B80_P20		
B70_P30		
B60_P40		

Figure A1. Effects of original and recycled PVB polymeric films with the blending of PVC and clay on the surface morphology obtained by optical microscopy.

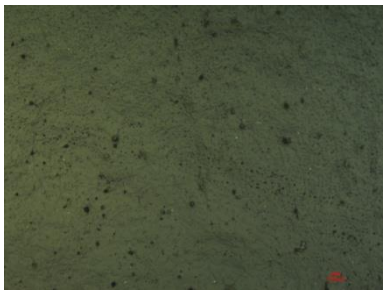
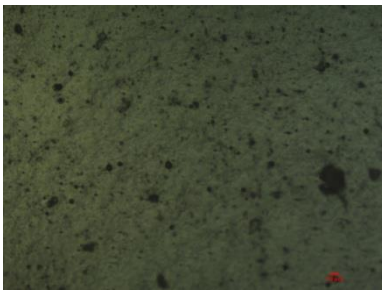
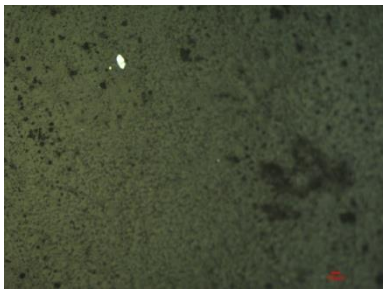
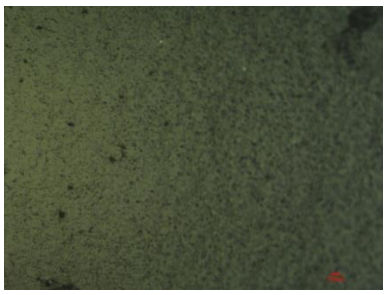
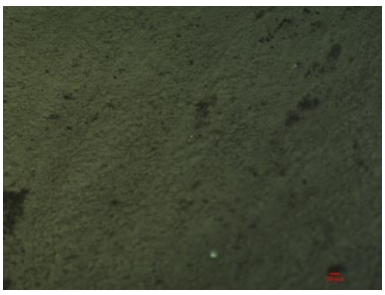
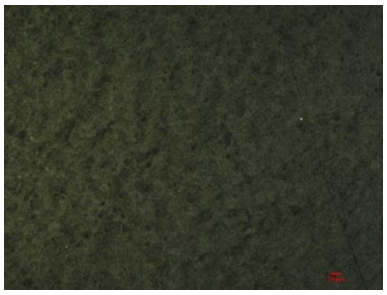
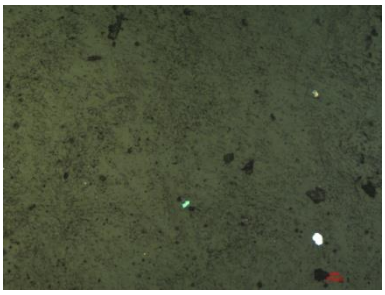
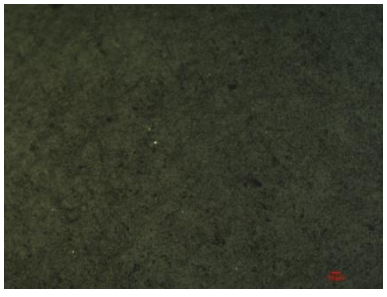
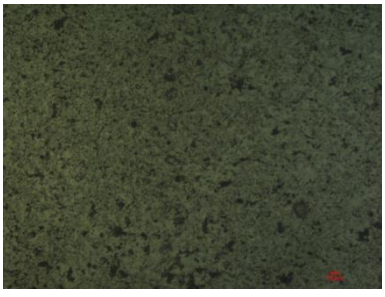
Sample Name	Original PVB	Recycled PVB
B50_P50		
B40_P60		
B30_P70		
B20_P80		
B10_P90		

Figure A1. Effects of original and recycled PVB polymeric films with the blending of PVC and clay on the surface morphology obtained by optical microscopy (continued).

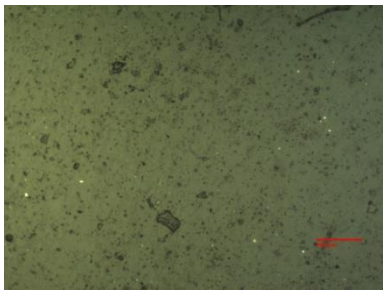
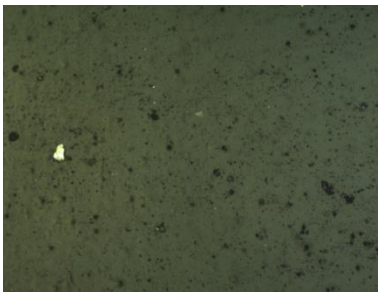
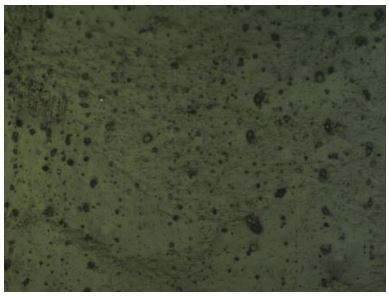
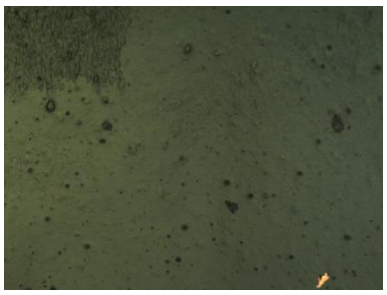
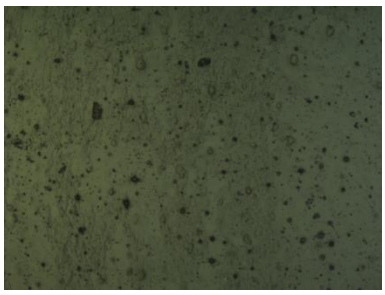
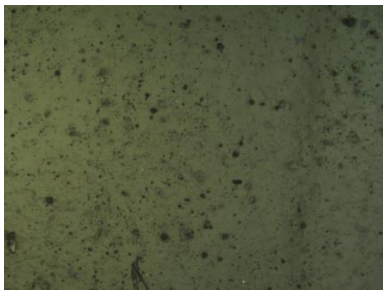
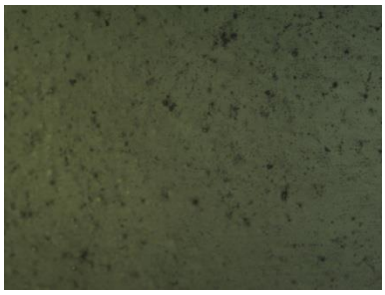
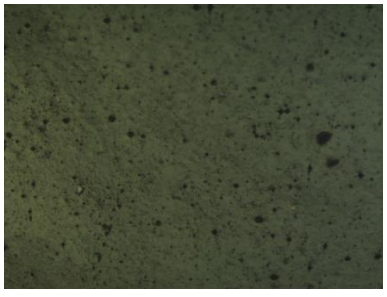
Sample Name	Original PVB	Recycled PVB
P100		**not available**
B90_P10_C0.1		
B90_P10_C1		
B95_C5		
B80_P20_C5		

Figure A1. Effects of original and recycled PVB polymeric films with the blending of PVC and clay on the surface morphology obtained by optical microscopy (continued).

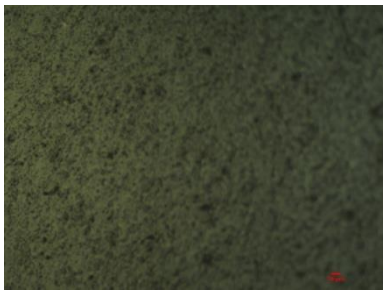
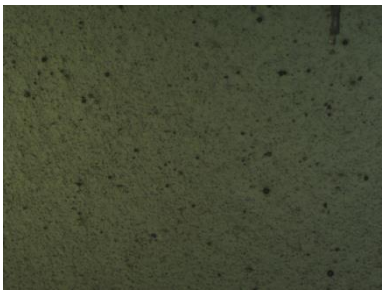
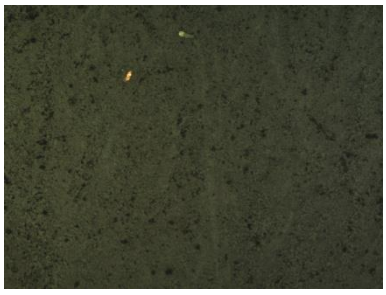
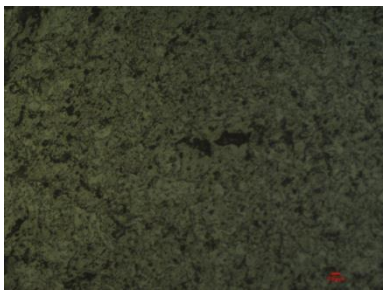
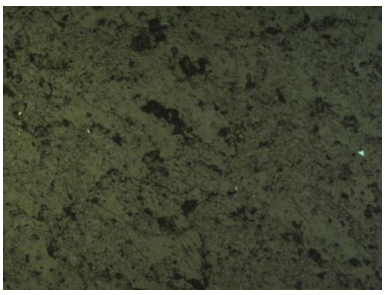
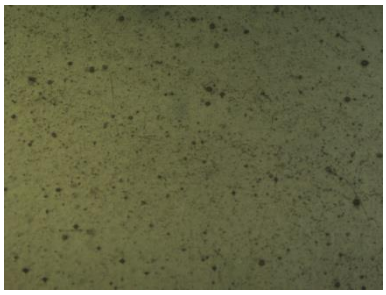
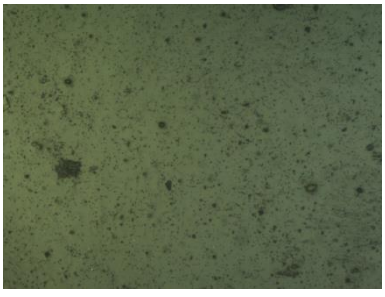
Sample Name	Original PVB	Recycled PVB
B60_P40_C5		
B40_P60_C5		
B20_P80_C5		
P95_C5		**not available**
B95_K5		

Figure A1. Effects of original and recycled PVB polymeric films with the blending of PVC and clay on the surface morphology obtained by optical microscopy (continued).

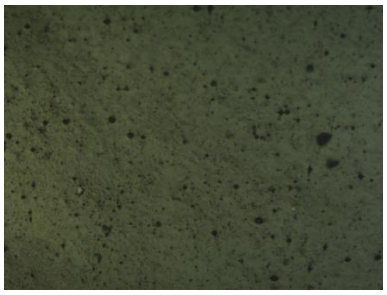
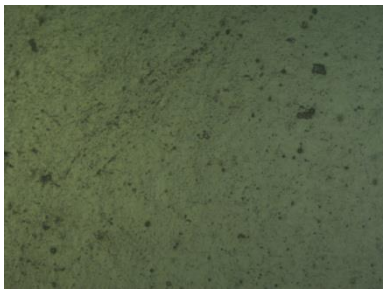
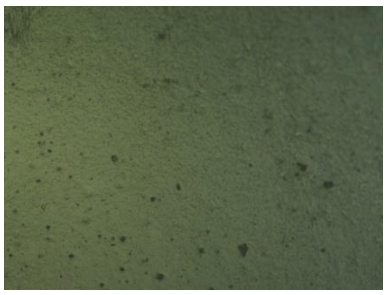
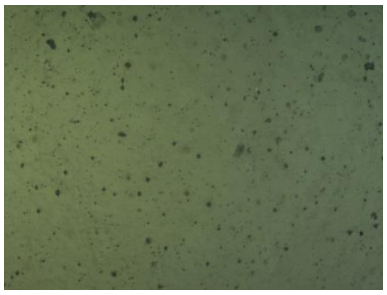
Sample Name	Original PVB	Recycled PVB
B20_P80_K5		**not available**
B40_P60_K5		**not available**
B60_P40_K5		**not available**
B80_P20_K5		**not available**

Figure A1. Effects of original and recycled PVB polymeric films with the blending of PVC and clay on the surface morphology obtained by optical microscopy (continued).

B. Mechanical test results for all polymeric films

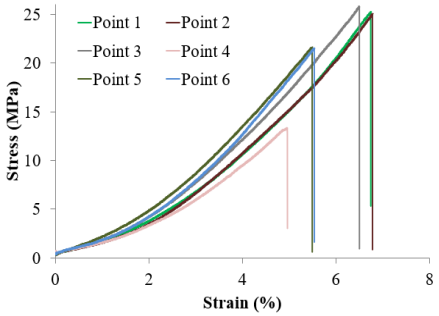
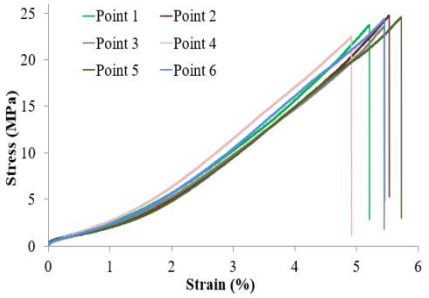
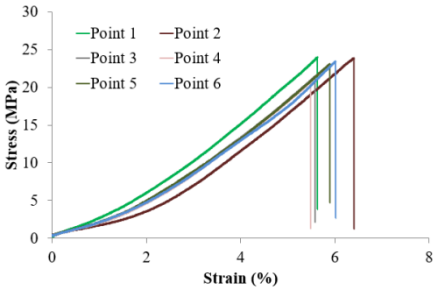
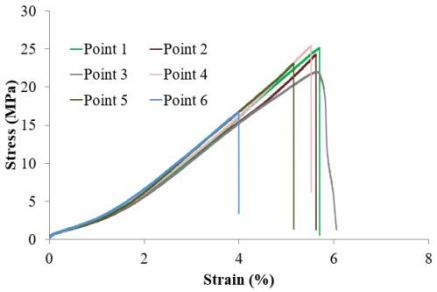
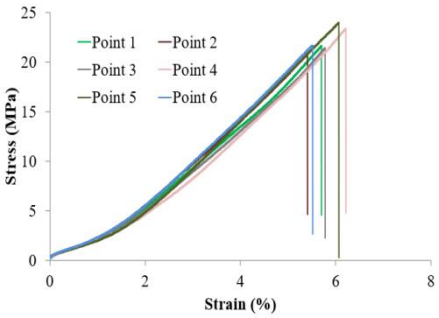
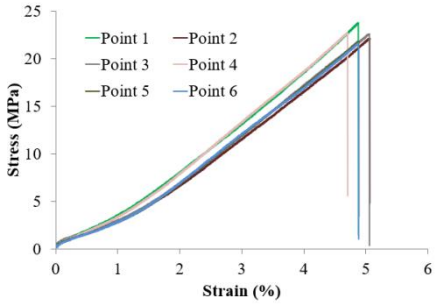
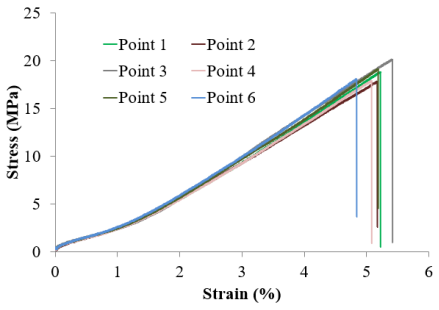
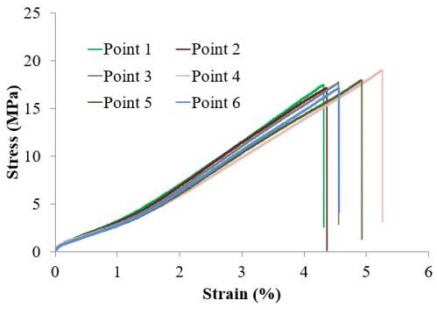
Sample Name	Original PVB	Recycled PVB
B100		
B90_P10		
B80_P20		
B70_P30		

Figure B1. The mechanical test results for the selected polymeric films.

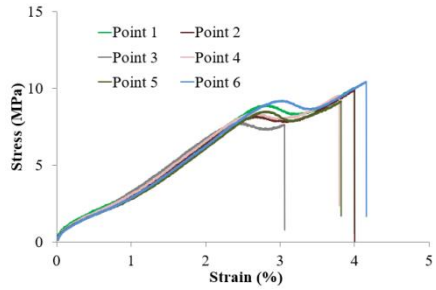
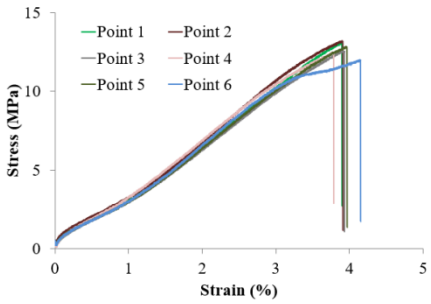
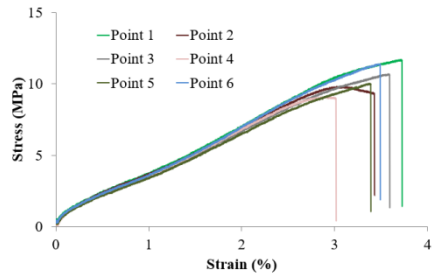
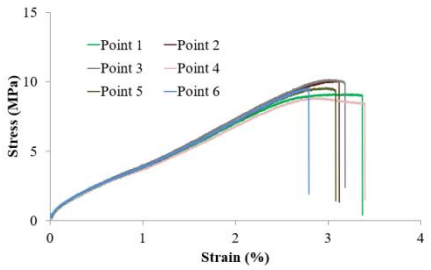
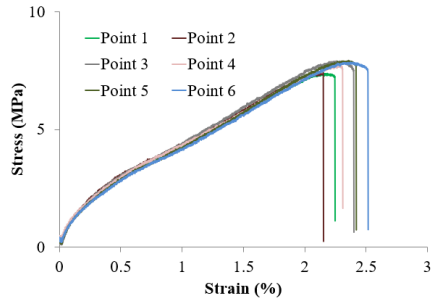
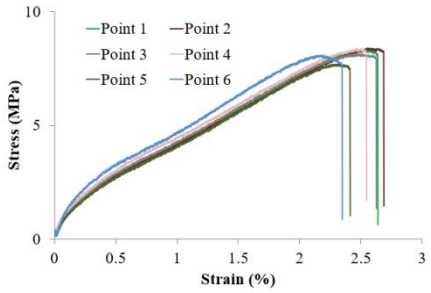
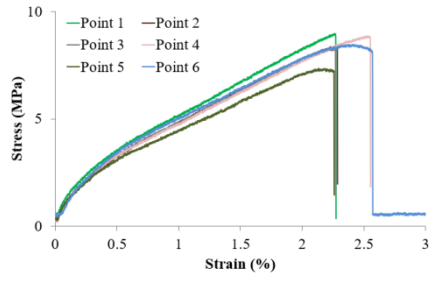
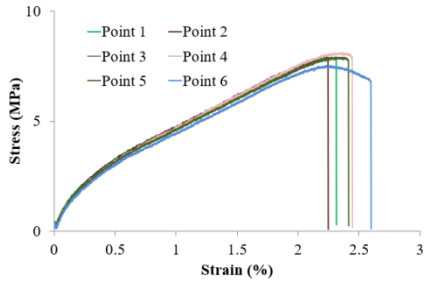
Sample Name	Original PVB	Recycled PVB
B60_P40		
B50_P50		
B40_P60		
B30_P70		

Figure B1. The mechanical test results for the selected polymeric films (continued).

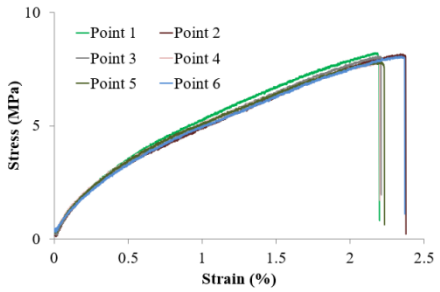
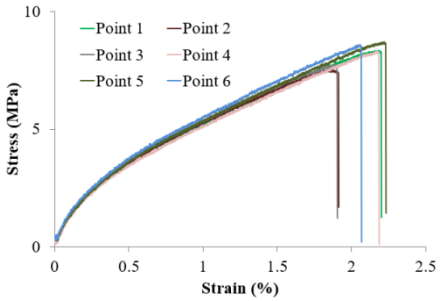
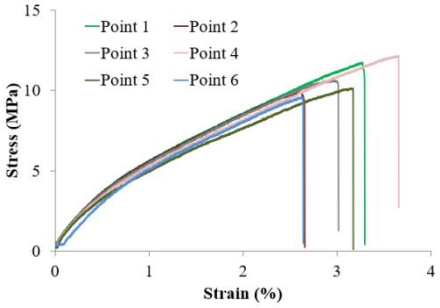
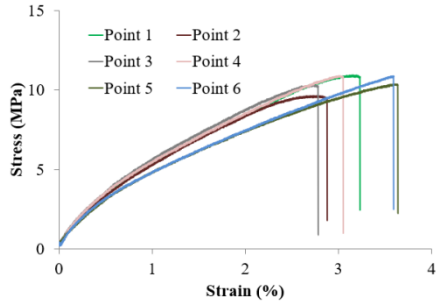
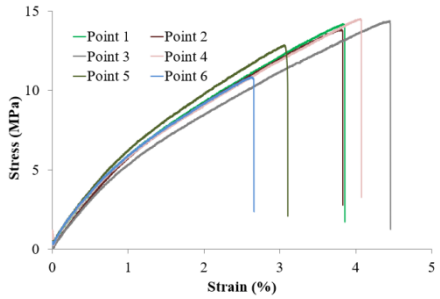
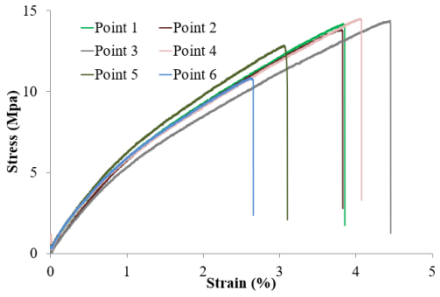
Sample Name	Original PVB	Recycled PVB
B20_P80		
B10_P90		
P100		not available
P95_C5		not available

Figure B1. The mechanical test results for the selected polymeric films (continued).

C. Thermal Degradation

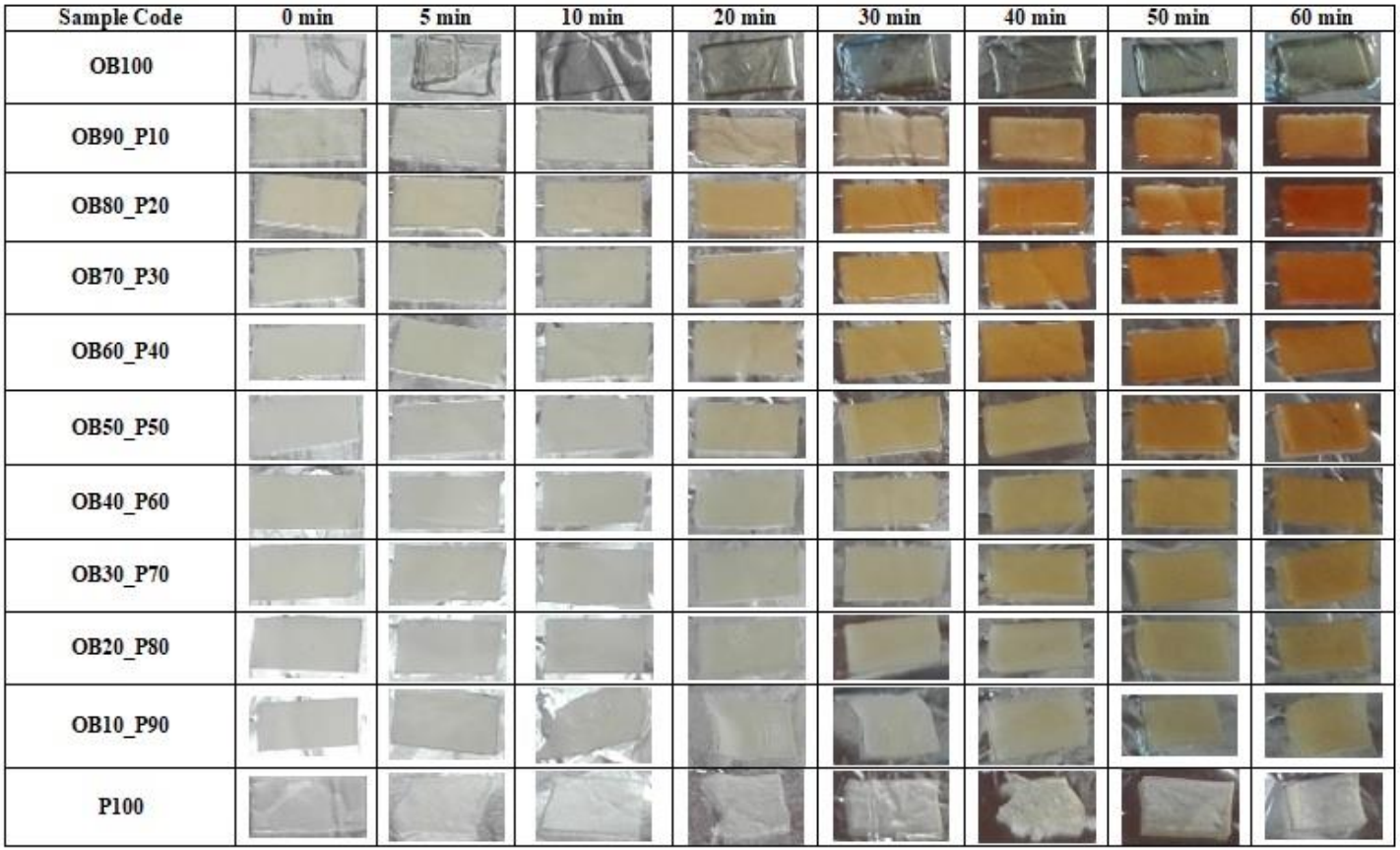


Figure C1. Color changes of OB/P polymeric films after thermal degradation.

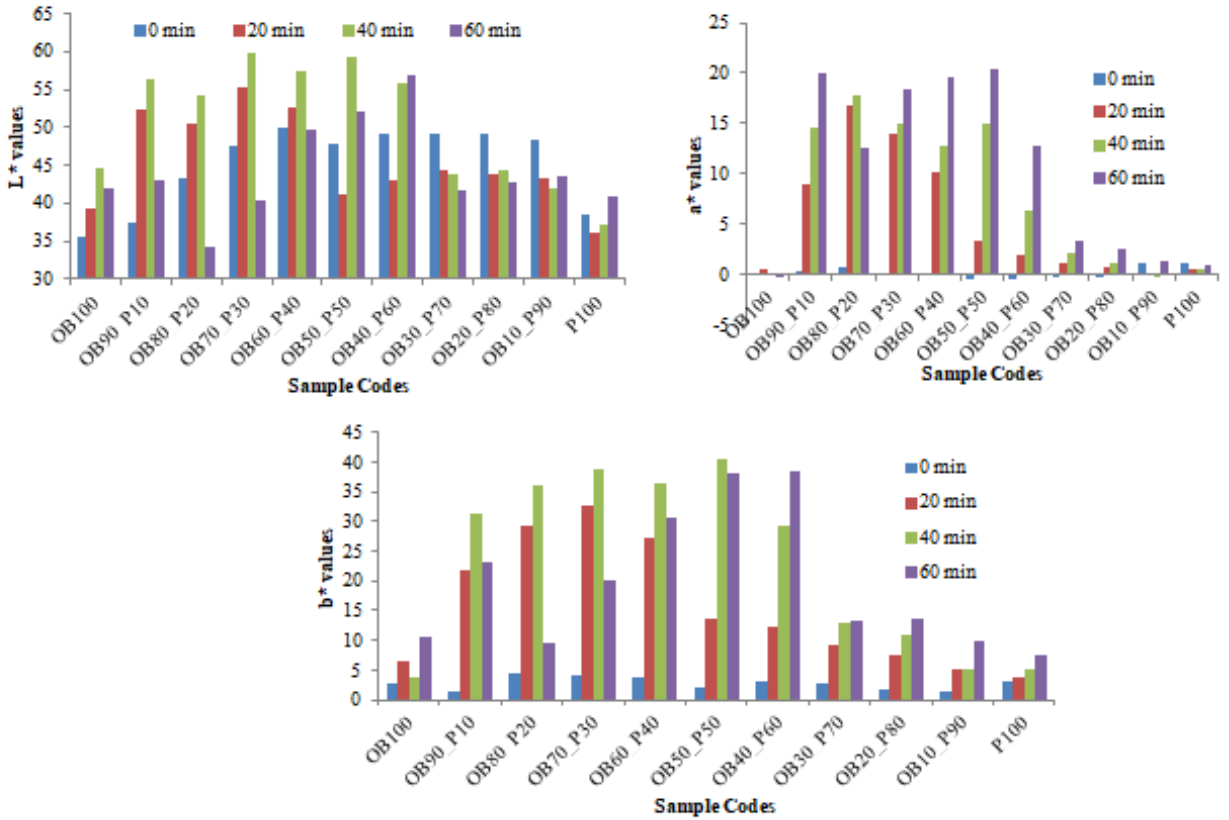


Figure C2. Comparison of L*, a* and b* values for OB/P samples.

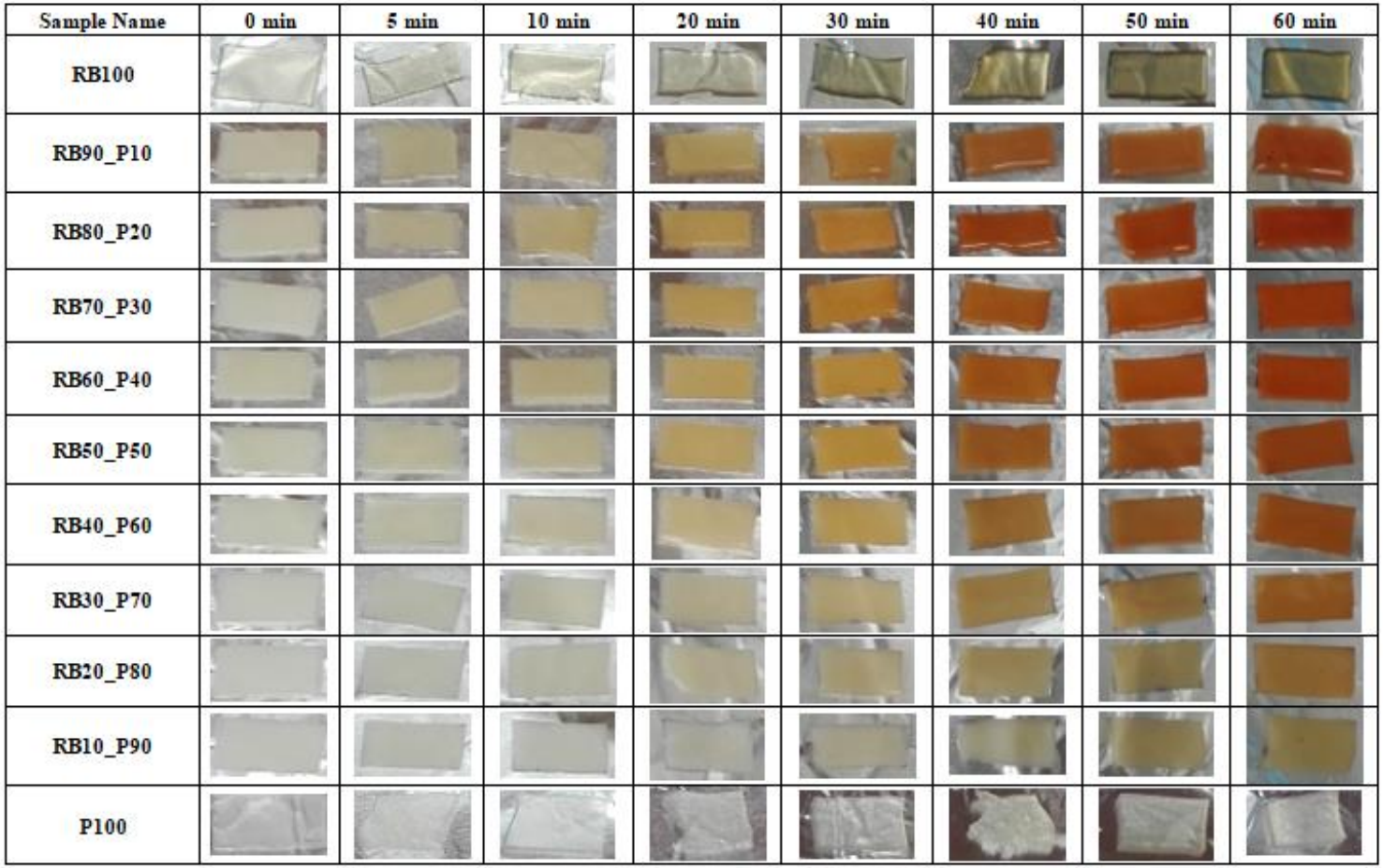


Figure C3. Color changes of RB/P polymeric films after thermal degradation

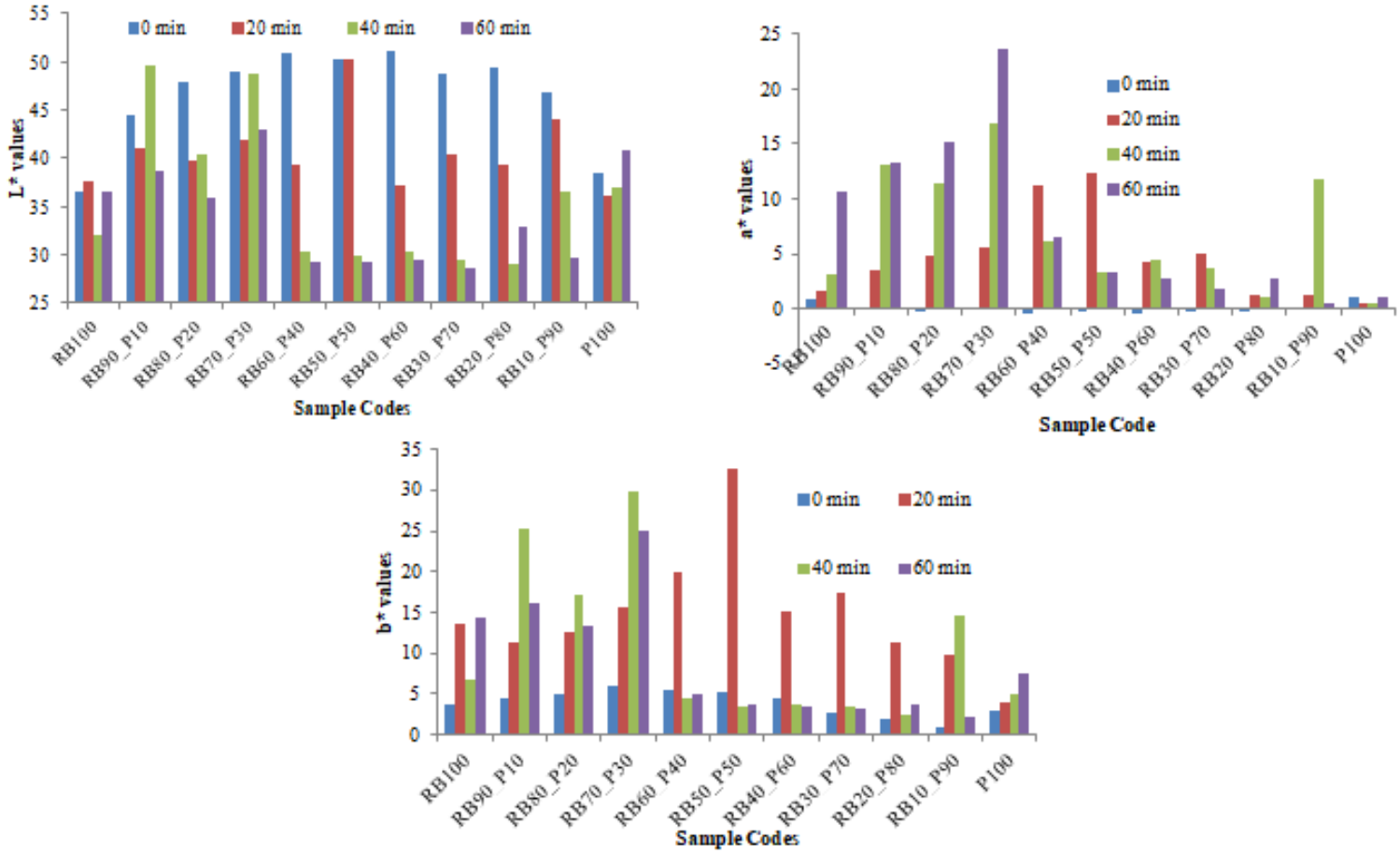


Figure C4. Comparison of L*, a* and b* values for RB/P samples.

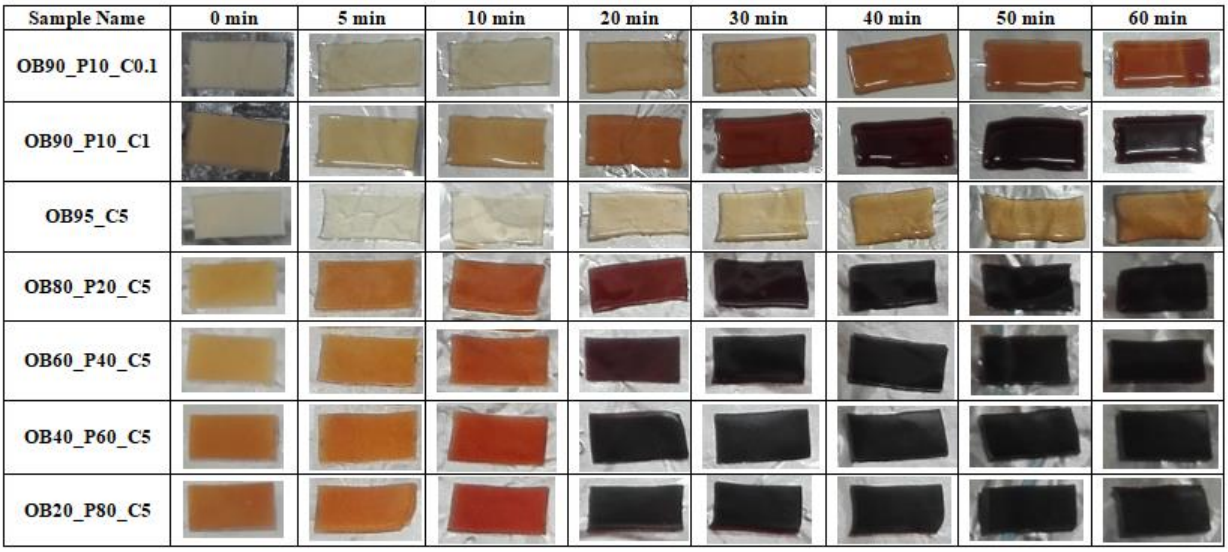


Figure C5. Color changes of OB/P and clay polymeric films after thermal degradation.

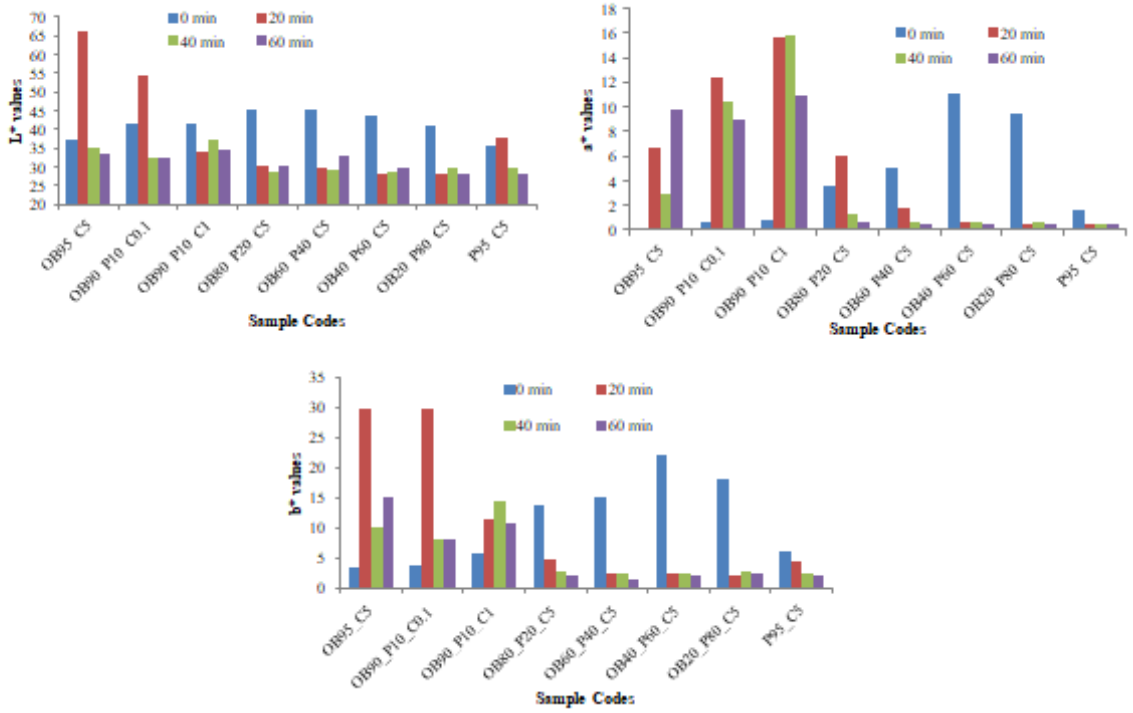


Figure C6. Comparison of L*, a* and b* values for OB/P/C and clay samples.

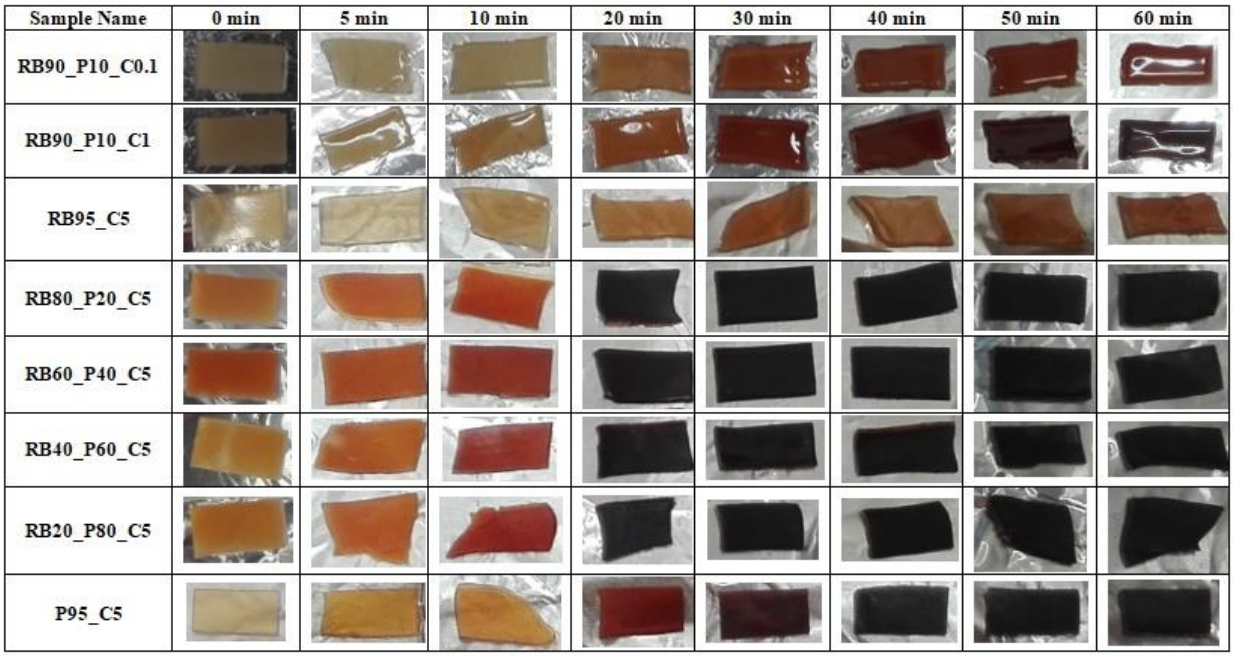


Figure C7. Color changes of RB/P and clay polymeric films after thermal degradation.

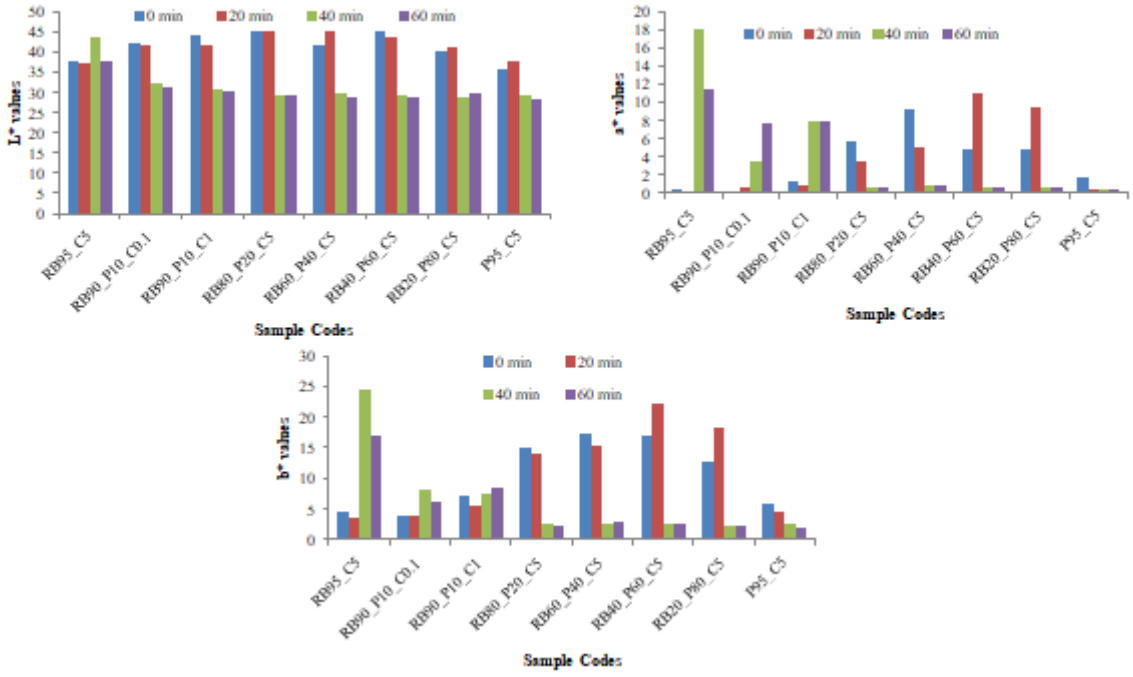


Figure C8. Comparison of L*, a* and b* values for RB/P samples with clay

Sample Name	0 min	5 min	10 min	20 min	30 min	40 min	50 min	60 min
OB95_K5								
RB95_K5								
OB80_P20_K5								
OB60_P40_K5								
OB40_P60_K5								
OB20_P80_K5								

Figure C9. Color changes of OP/P and calcium carbonate samples after thermal degradation.

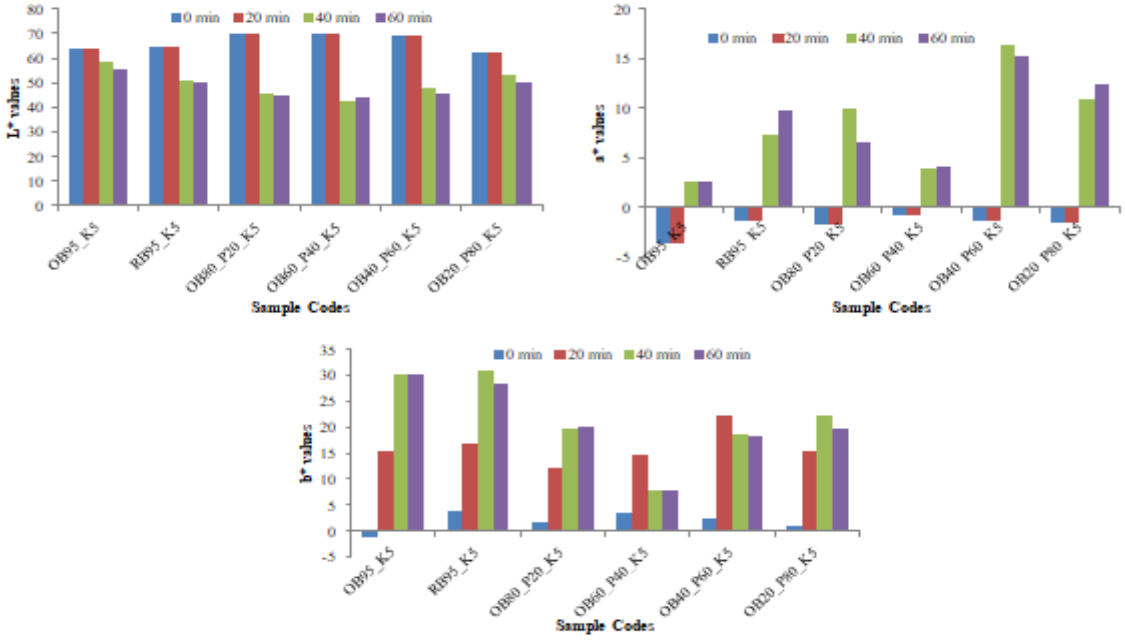


Figure C10. Comparison of L*, a* and b* values for OB/P/K5 sample.

D. X-Ray Diffraction (XRD) Analysis

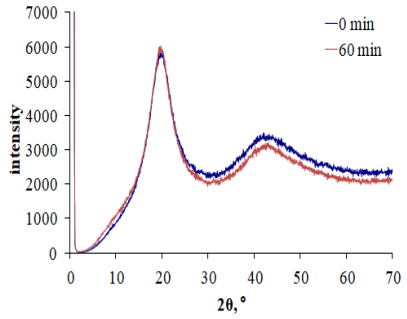
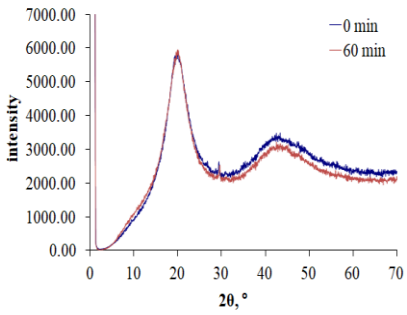
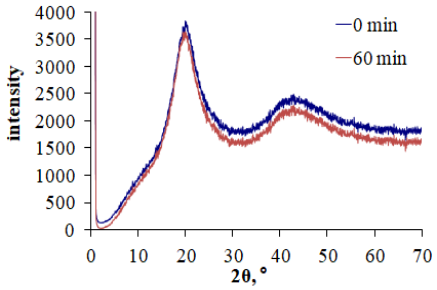
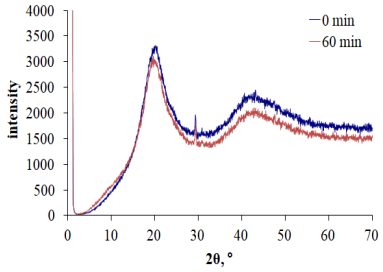
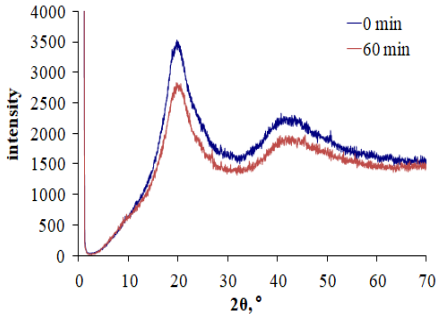
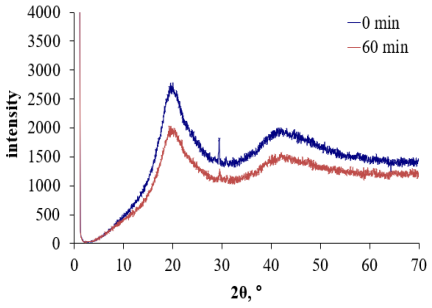
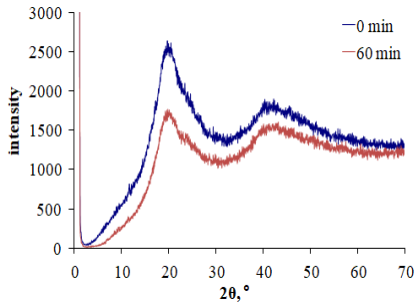
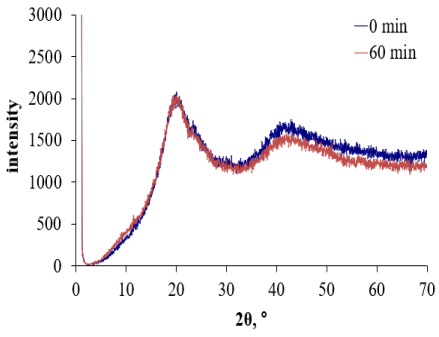
Sample Name	Original PVB	Recycled PVB
B100		
B90_P10		
B80_P20		
B70_P30		

Figure D1. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition at 0 minutes and 60 minutes during thermal degradation.

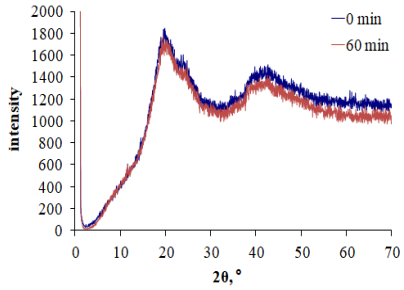
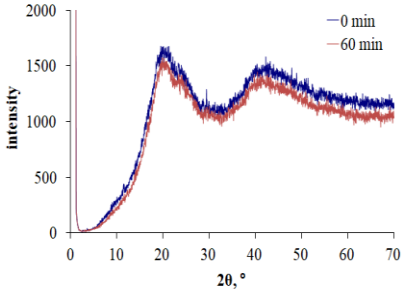
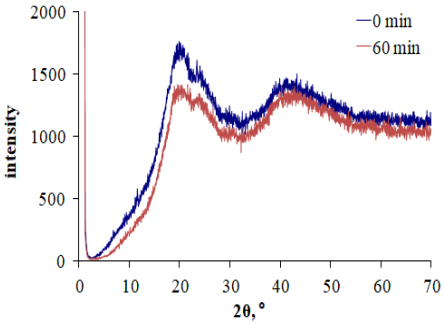
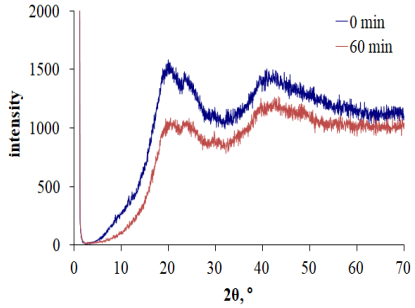
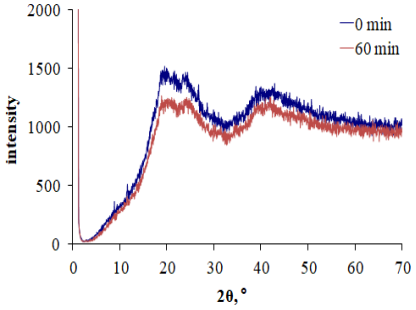
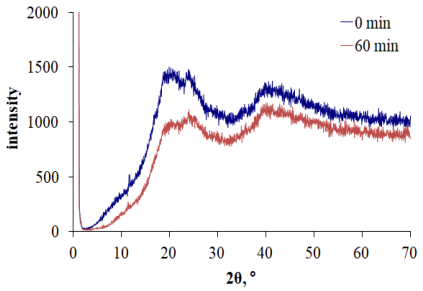
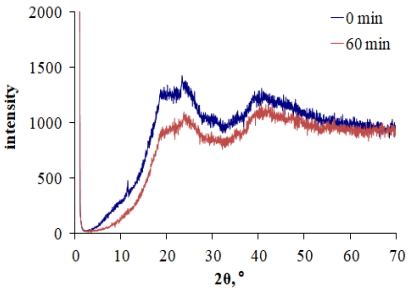
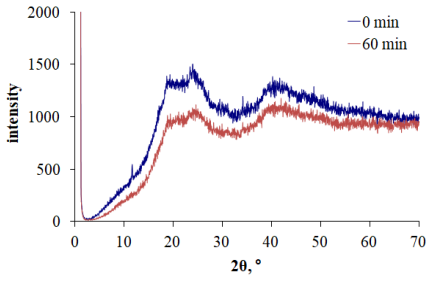
Sample Name	Original PVB	Recycled PVB
B60_P40		
B50_P50		
B40_P60		
B30_P70		

Figure D1. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition at 0 minutes and 60 minutes during thermal degradation (continued).

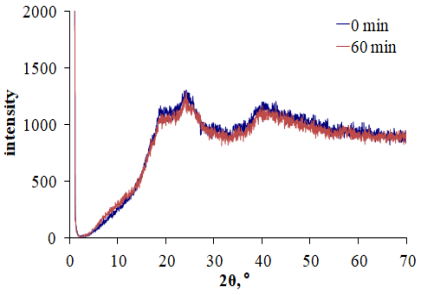
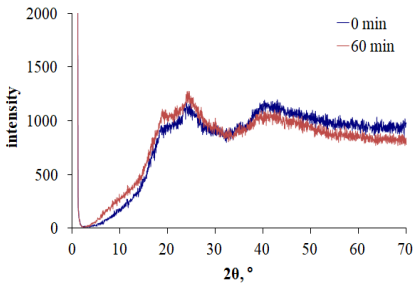
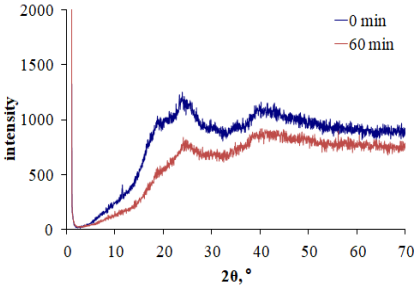
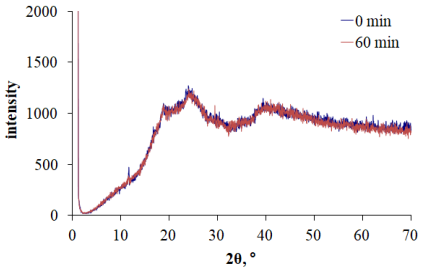
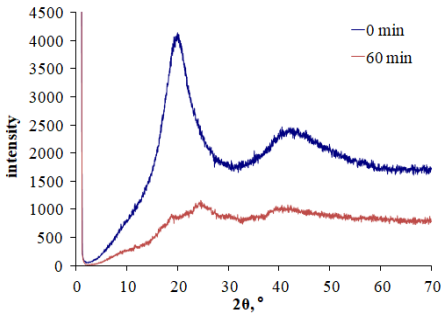
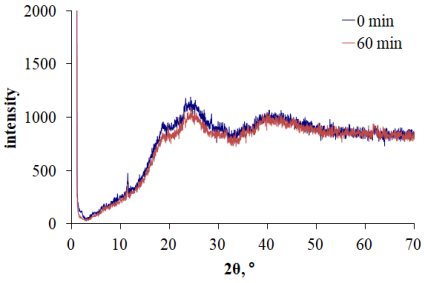
Sample Name	Original PVB	Recycled PVB
B20_P80		
B10_P90		
P100		not available
P95_C5		not available

Figure D1. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition at 0 minutes and 60 minutes during thermal degradation (continued).

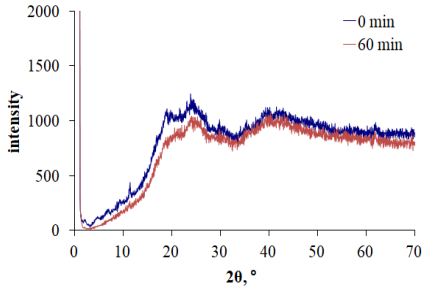
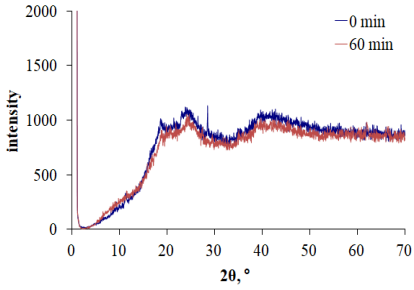
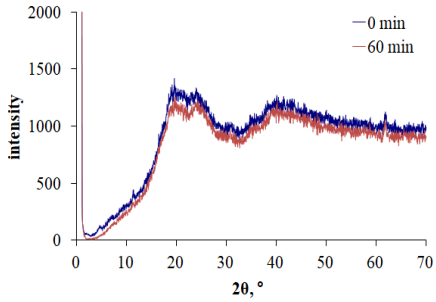
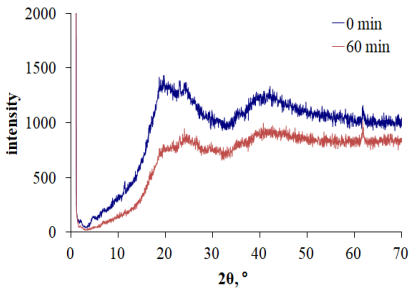
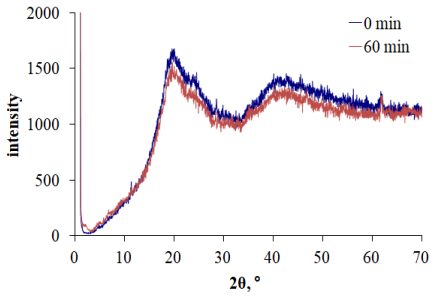
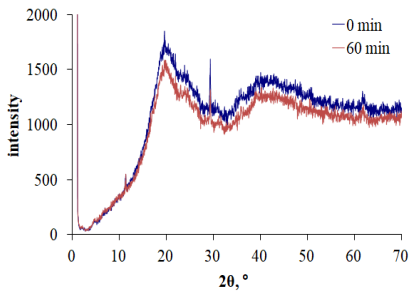
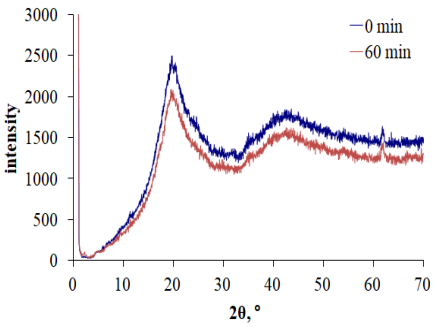
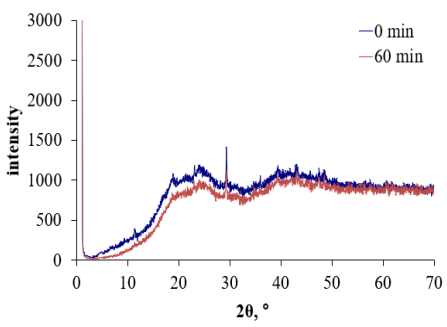
Sample Name	Original PVB	Recycled PVB
B20_P80_C5		
B40_P60_C5		
B60_P40_C5		
B80_P20_C5		

Figure D1. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition at 0 minutes and 60 minutes during thermal degradation (continued).

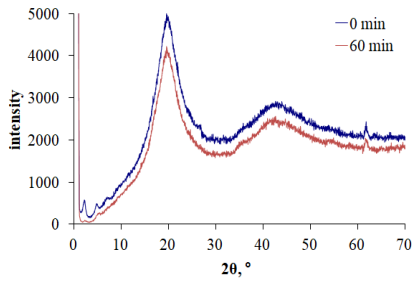
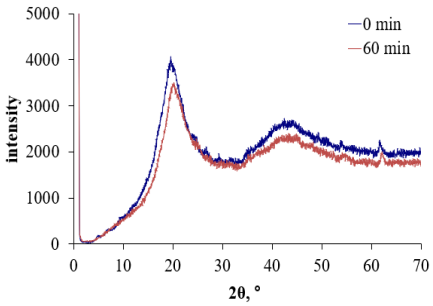
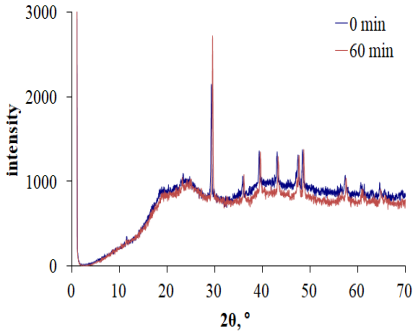
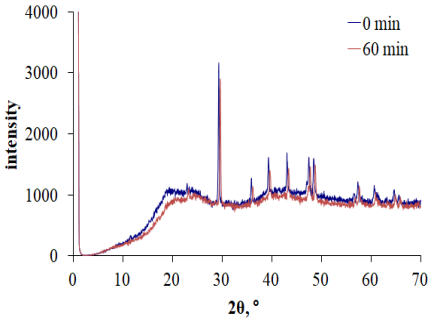
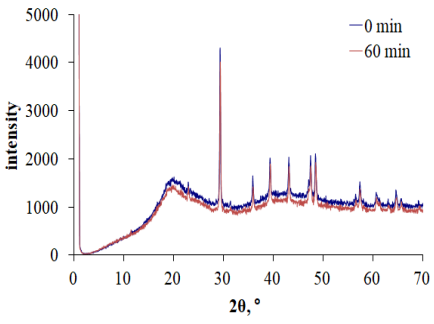
Sample Name	Original PVB	Recycled PVB
B95_C5		
B20_P80_K5		not available
B40_P60_K5		not available
B60_P40_K5		not available

Figure D1. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition at 0 minutes and 60 minutes during thermal degradation (continued).

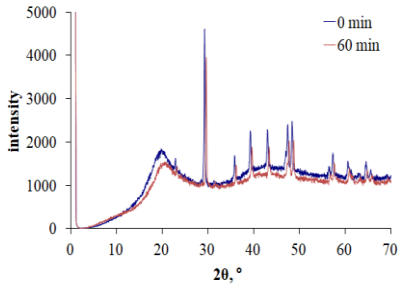
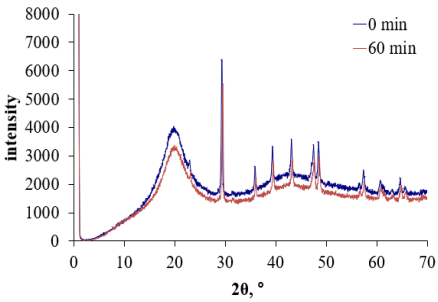
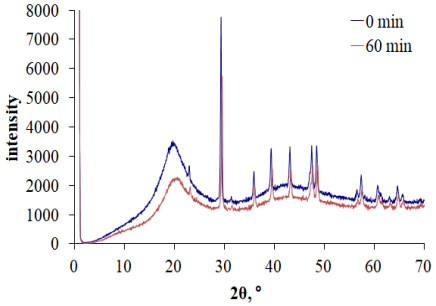
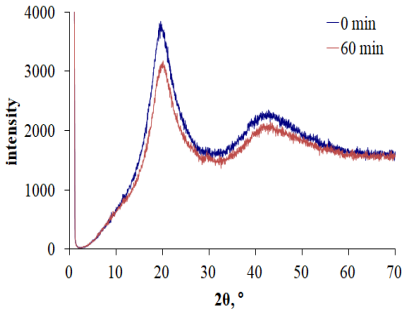
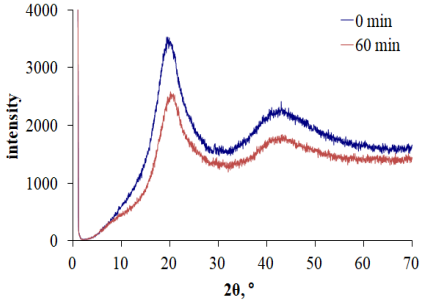
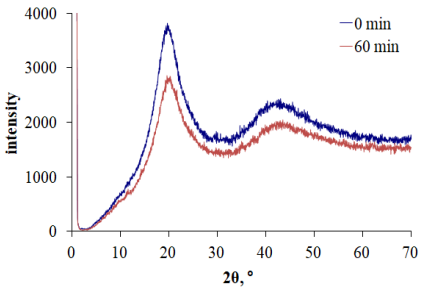
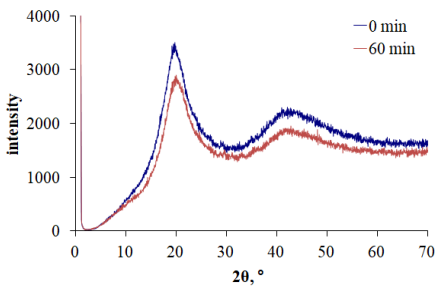
Sample Name	Original PVB	Recycled PVB
B80_P20_K5		not available
B95_K5		
B90_P10_C0.1		
B90_P10_C1		

Figure D1. XRD results of original and recycled PVB with PVC, Clay and Calcium Carbonate addition at 0 minutes and 60 minutes during thermal degradation (continued).