Óbuda University
Doctoral School of Materials Science and Technologies

Neumann János Egyetem

Effect of Reprocessing of Polyester/ Montmorillonite Nanocomposites

Prepared by

Zoubeida Taha Taha

PhD. student

Supervisors

Dr. Andrea Ádámné Major,

Prof. Ferenc Ronkay

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The problem to be solved

Unknown properties of recycled nanocomposites.

Aim of the research

- Study the **effect of recycling** on the morphology, mechanical and thermal properties of MMT-nanocomposites with different polyester matrices.

Practical part

Types of polyesters

Petroleum-based

- Polyethylene terephthalate (PET)
- Polybutylene Terephthalate (PBT)

Bio-based

- Polybutylene succinate (PBS)
- Polylactic acid (PLA)

Montmorillonite

Practical part

Test samples

1- After drying I compounded the nanocomposites added 6 wt.% of MMT to each of PET, PBS, PBT and PLA using a twin screw extruder (1x extr.)

2- I repeated the extrusion for the reprocessed samples (2x extr.)

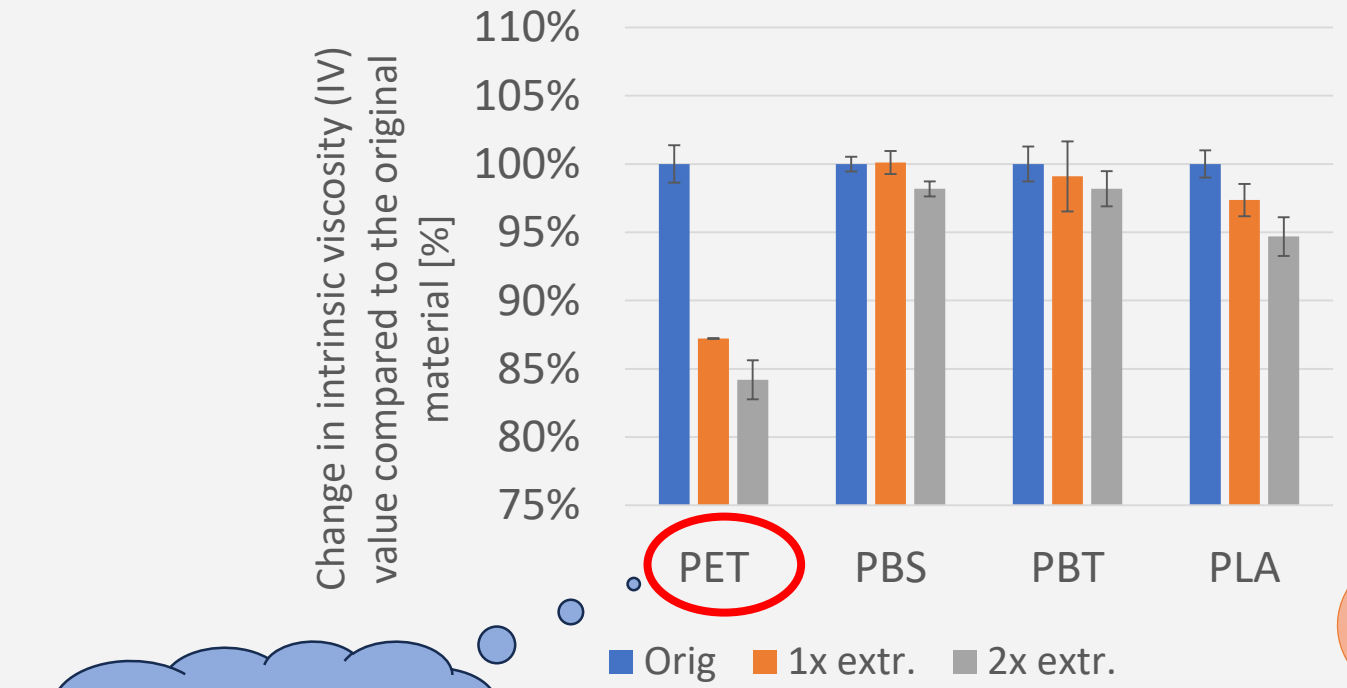
3- I dried all nanocomposite materials in an oven.

4- I used injection molding to make the test samples.

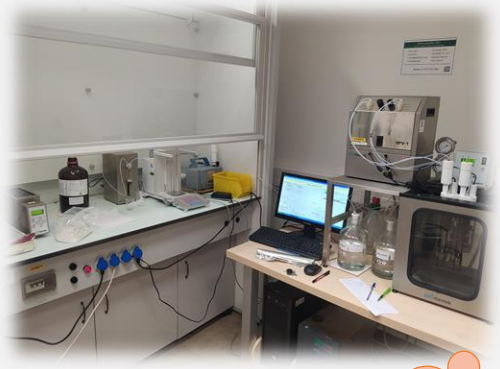
Practical part

Intrinsic viscosity (IV)

- Effect of recycling on the molecular weight of PET, PBS, PBT and PLA.



The highest degradation



Mark-Houwink

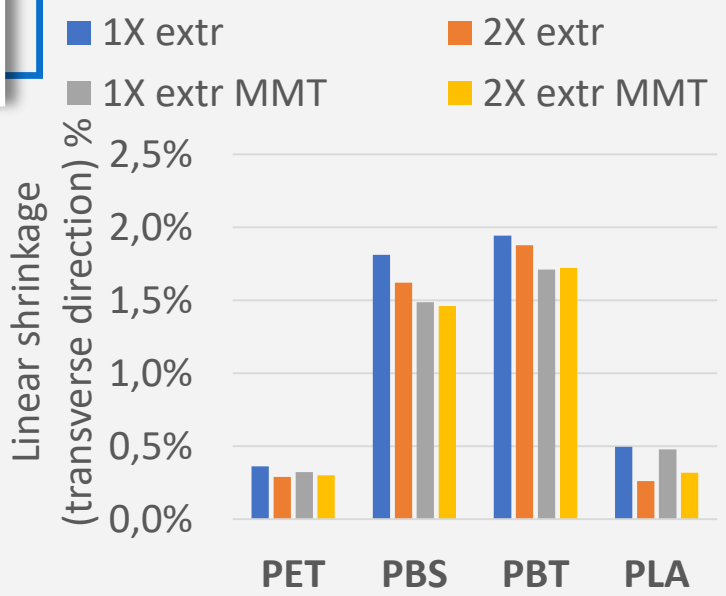
$$[\eta] = K M^{\alpha}$$

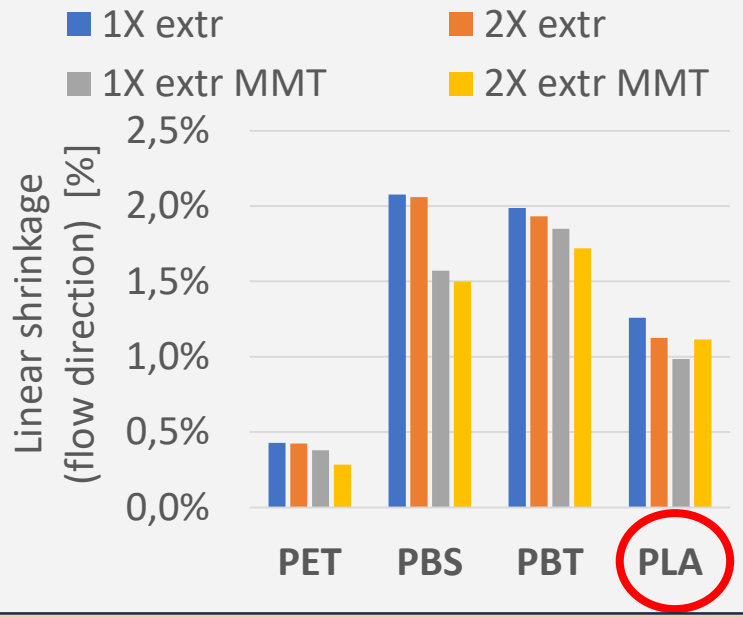
η: Intrinsic viscosity.
M: molecular weight.
K, α: constants.

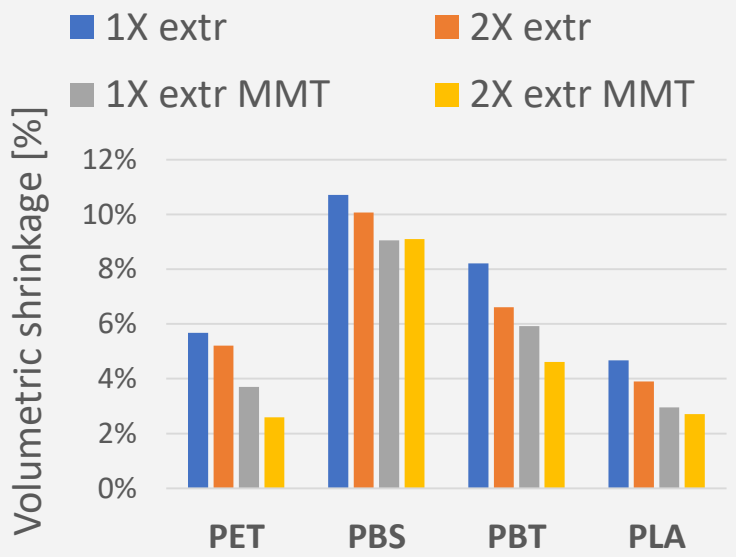
Practical part

Shrinkage of composites (PET, PBS, PBT, PLA / MMT)

- The effect of adding MMT and recycling on the shrinkage of composite materials after 1 day.





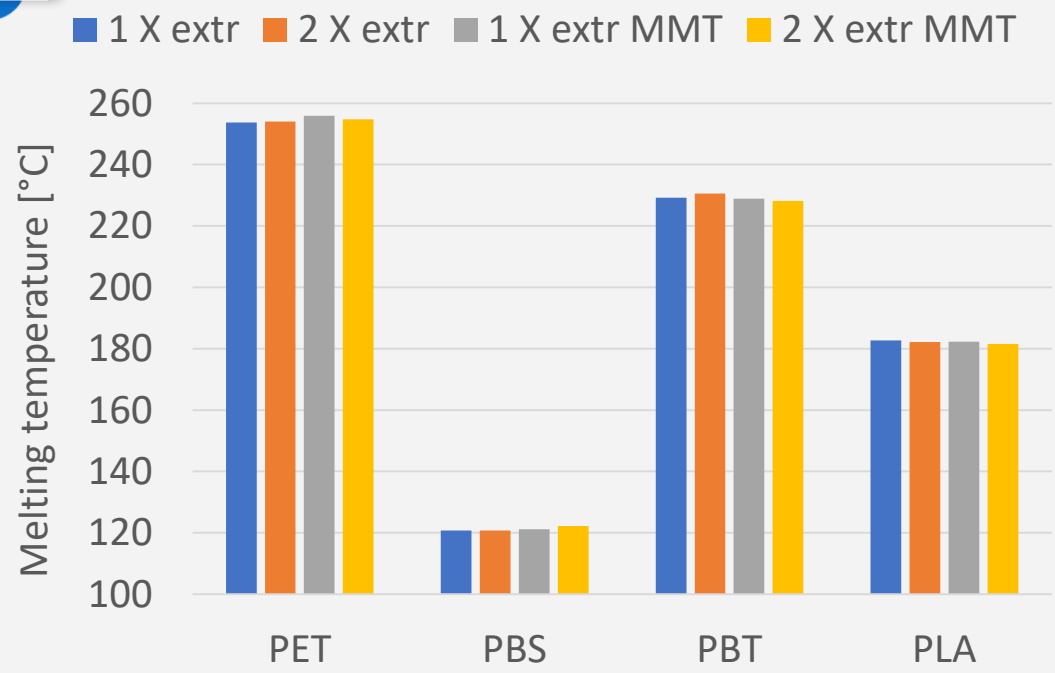


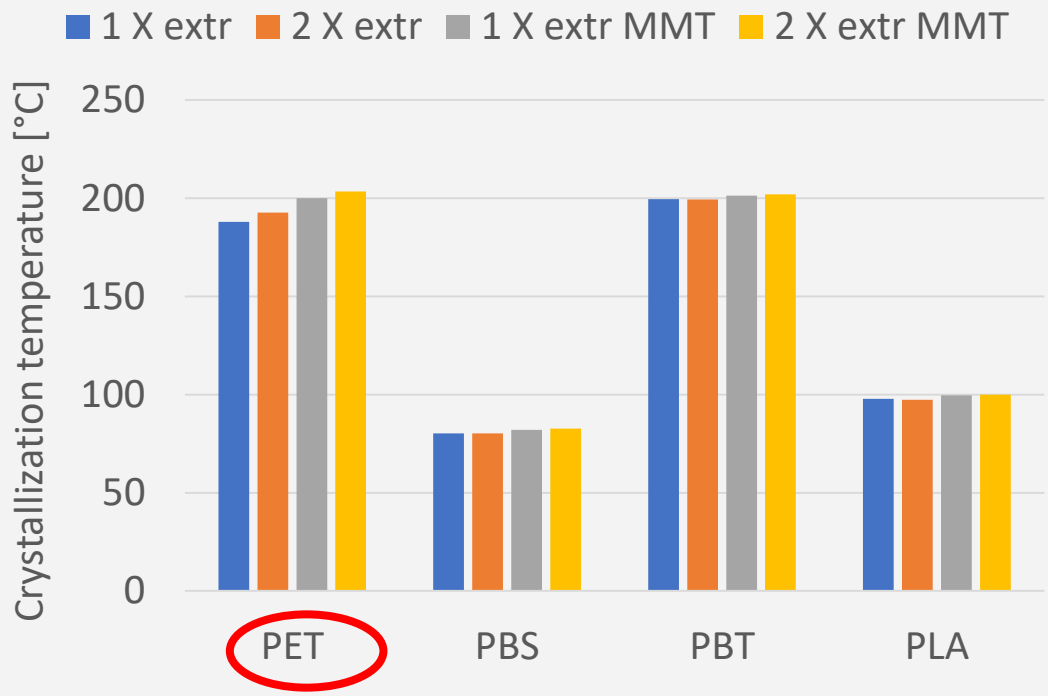
- Shrinkage variation indicates anisotropy.
- 2X extr → Shrinkage ↓ of all polyesters.
- Adding MMT → shrinkage ↓
- 2X extr MMT → shrinkage **decreases,**
- exception **PLA** (FD) $T_m < T_{recommended}$ → Strong molecular orientation → higher relaxation of the chains → higher shrinkage.

Practical part

Non-Isothermal crystallization of PET, PBS, PBT and PLA

I scanned them by using a DSC during the following cooling rates 2.5, 5, 10, 20, 40°C/ min to determine the crystallization and melting temperature.



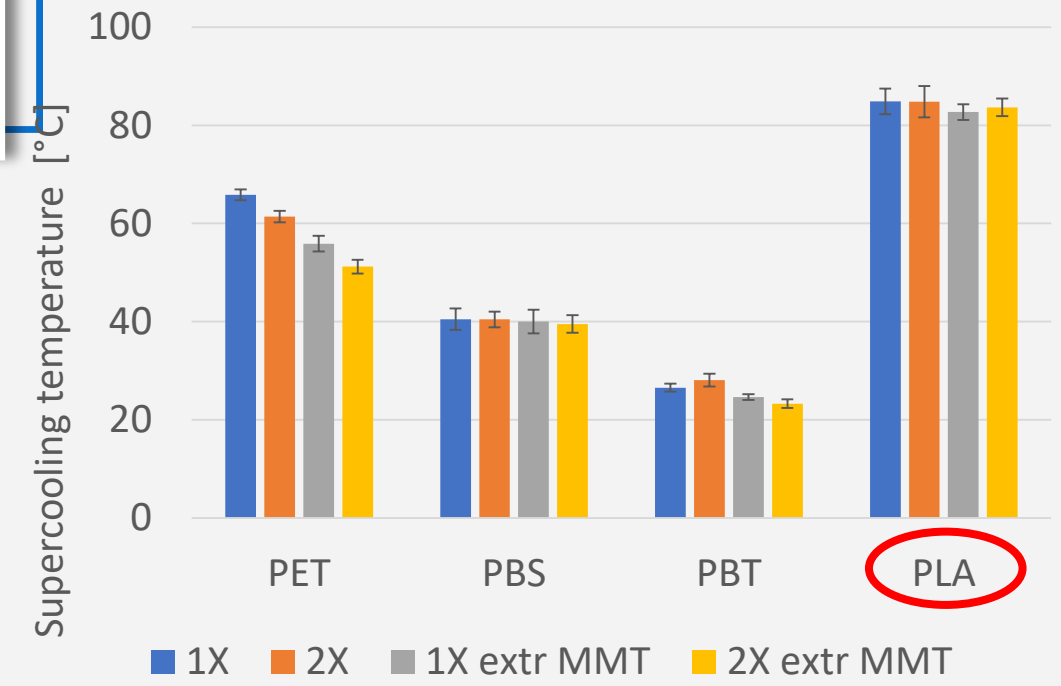


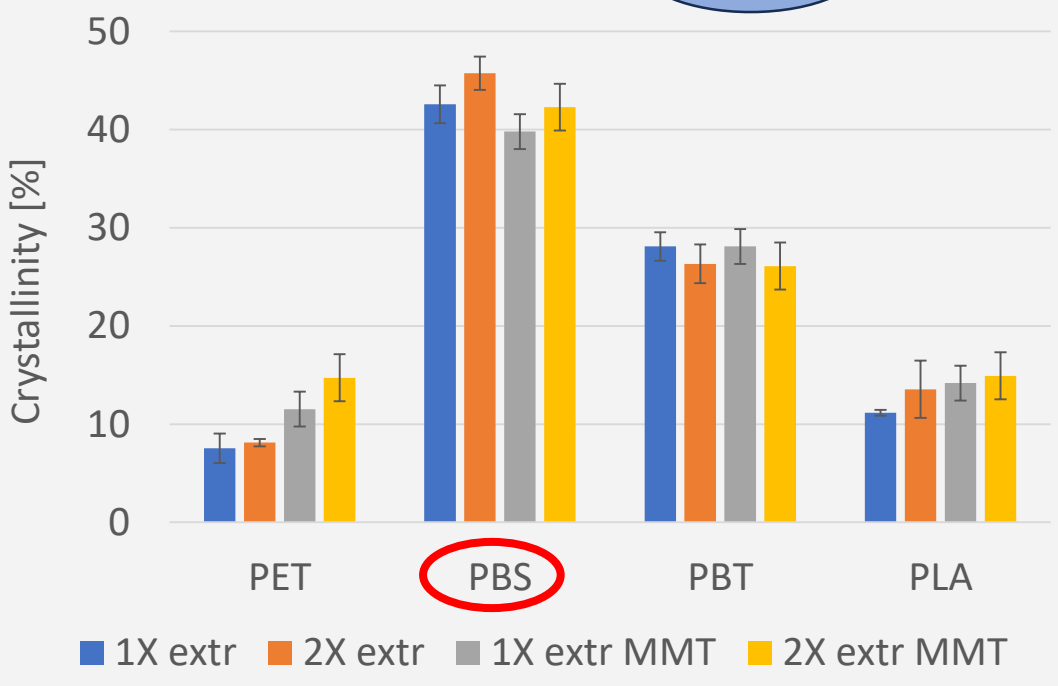
- T_m: **Φ** change significantly with reprocessing and MMT.
- T_c → By reprocessing → **Φ** change, except **PET**↑ :Low viscosity so more chain order.
  → By adding MMT → ↑ of all polyesters.

Practical part

Crystallinity of composites PET, PBS, PBT and PLA

Supercooling ↔ CRF



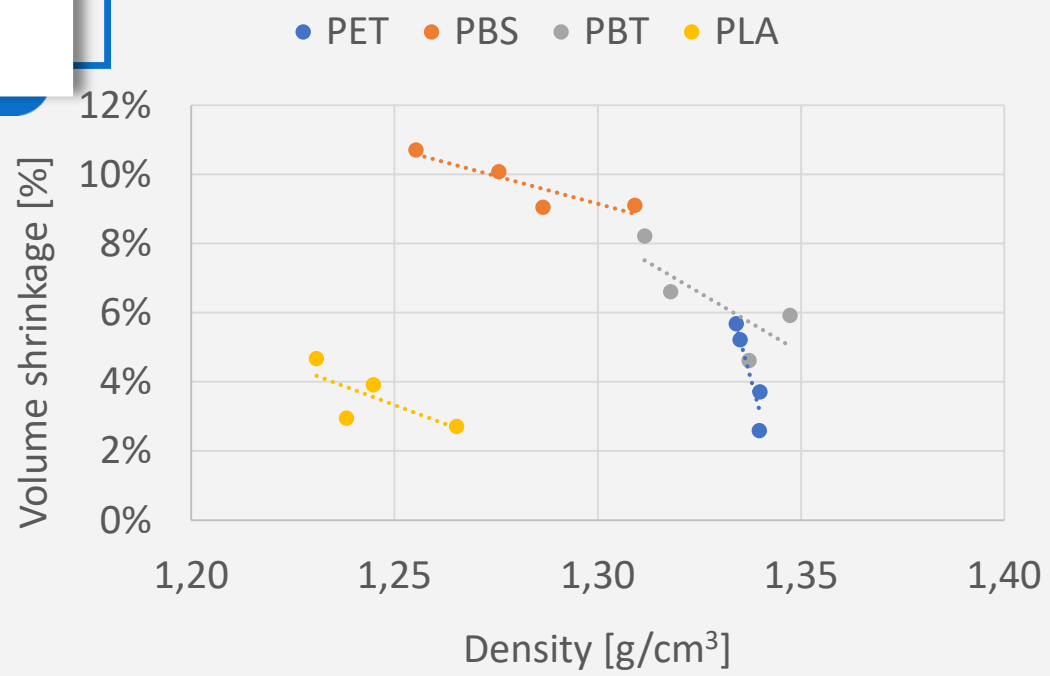


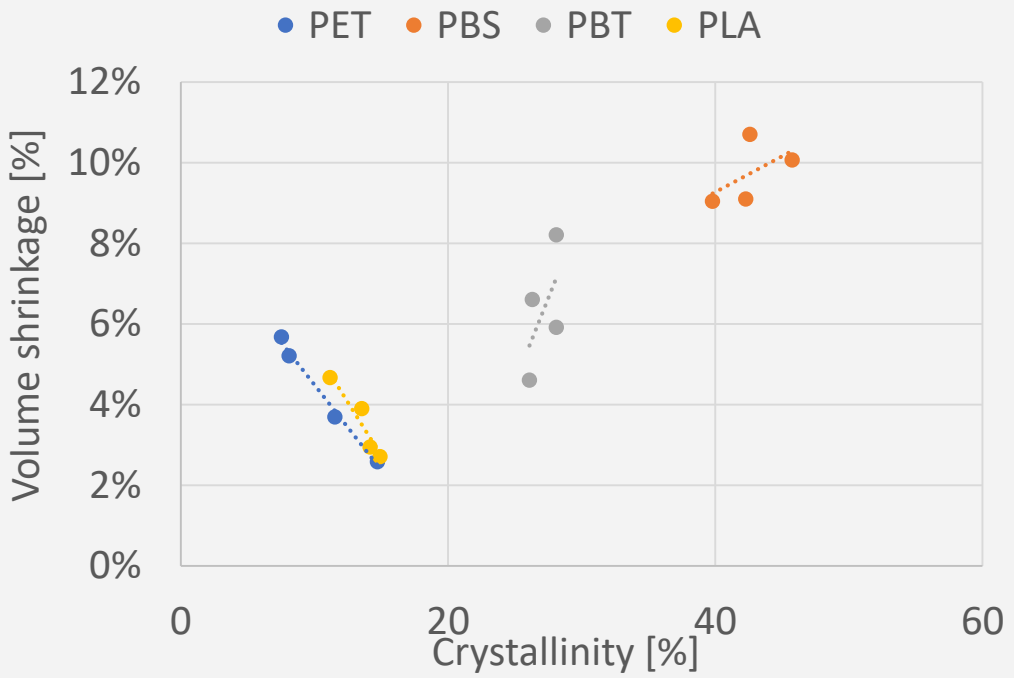
- Supercooling temperature = T_m - T_c.
- It depends on the type of polyester: **PLA** is the highest , higher cooling rate.
- $T_{Supercooling}$ → By reprocessing: ↓**PET**, ↑**PBT, Φ PLA and PBS** .
  → By adding MMT → ↓ in all cases, **except** for **PBS,** Prevent the order of chains.
- **PBT** and **PBS** have higher crystallization rates due to higher molecular mobility**.**

Practical part

Relationship between volumetric shrinkage, density and crystallinity.

I calculated the density of the samples after 1 day.





- Reprocessing and the presence of MMT have an effect on the <u>density</u> after shrinkage.
- Negative correlation: Structural changes ➡ reduce the space available ➡ decrease shrinkage.
- There is a relationship between volumetric shrinkage and crystallinity, especially for **PET** and **PLA**.
- Relaxation of the recoverable elastic strain ➡ restricts the movement of amorphous regions.

Practical part

Scanning Electron Microscope (SEM) - Energy Dispersive Spectroscopy (EDS) analysis of composites (PET, PBS, PBT, PLA / MMT)

I scanned the fracture surface of the samples after Izod impact test by using EDS.

10-20 µm

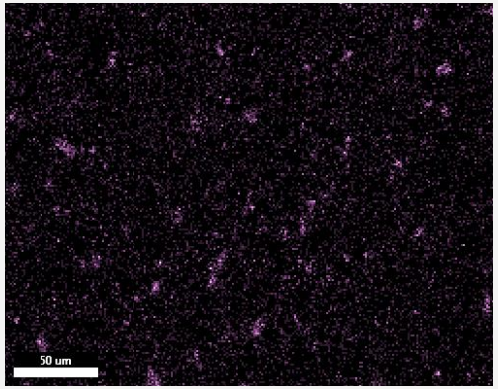
Si- MMT- 1X

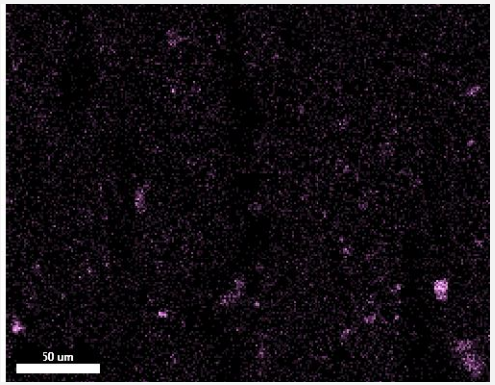
Si- MMT- 2X

Si- MMT- 1X

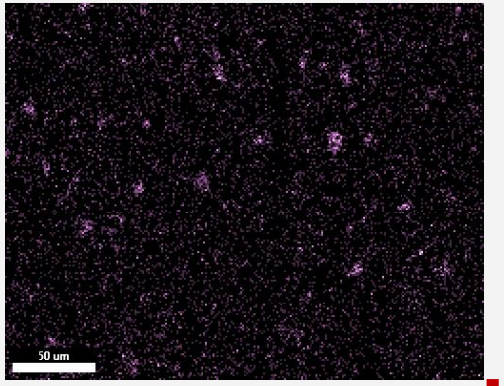
Si- MMT- 2X

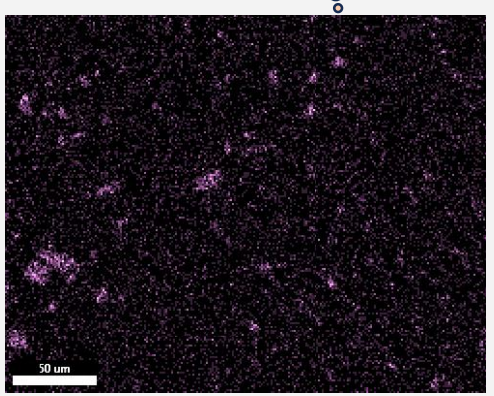
PBT





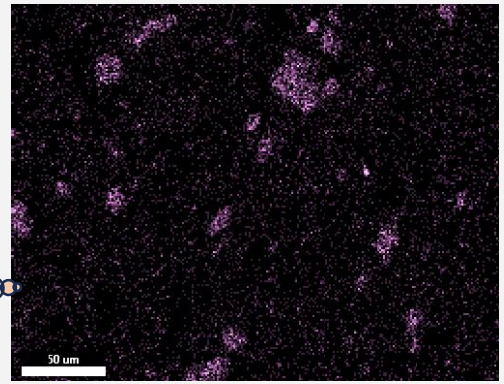
PET

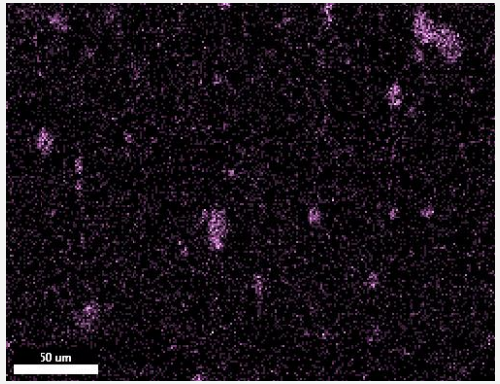




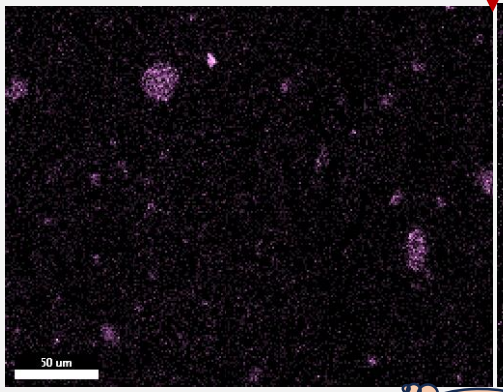
5-10 µm

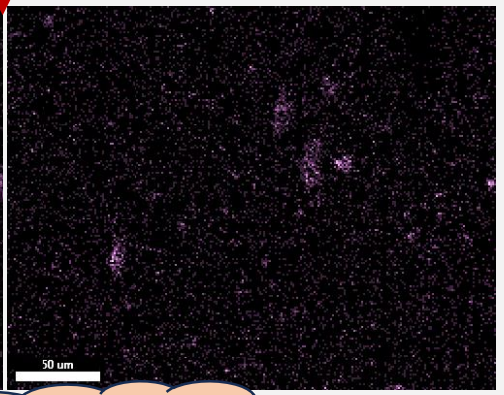
PBS





PLA





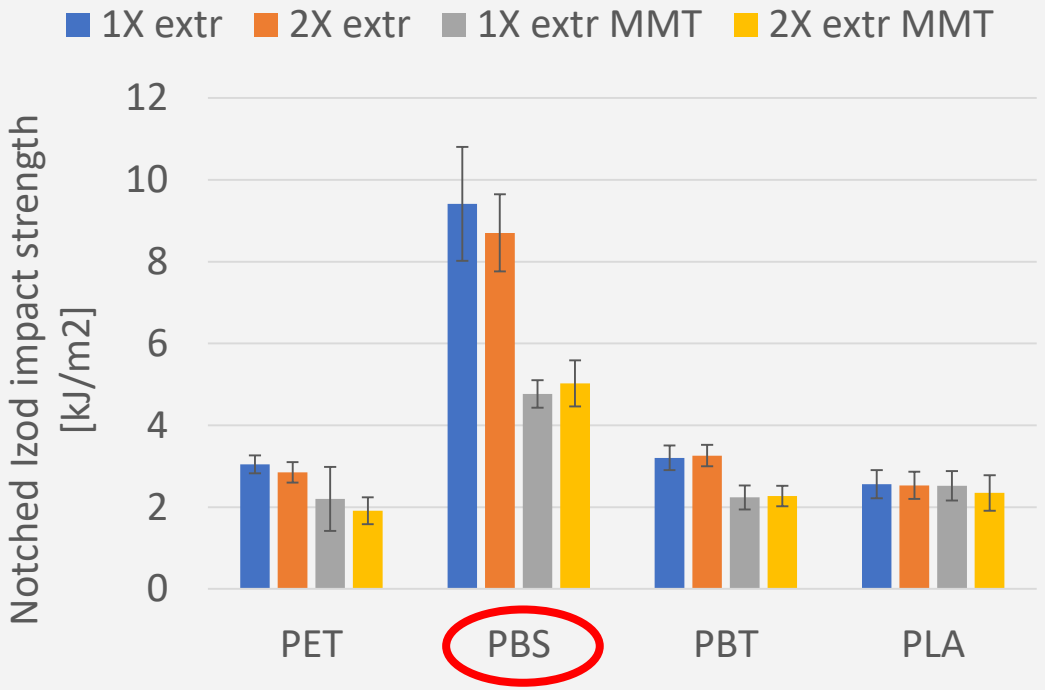
10-50 µm

10-30 µm

Practical part

Izod impact of composites (PET, PBS, PBT, PLA / MMT)

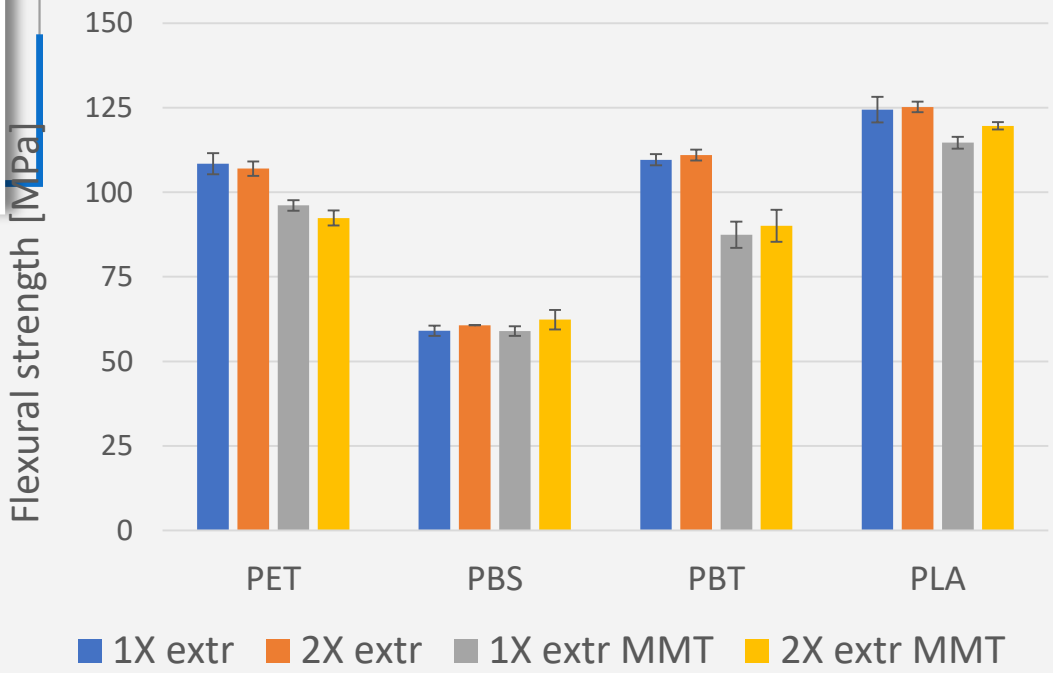
The effect of adding MMT and recycling on Izod impact strength of composite materials.

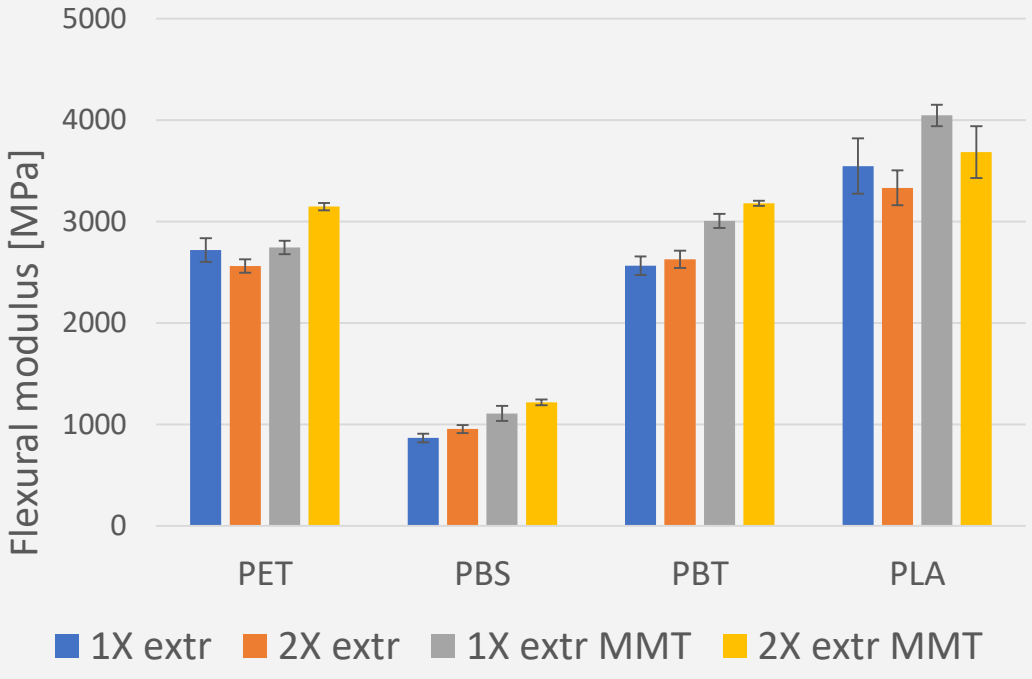


- **PBS** is the **highest** impact strength, **PET** and **PLA** are the lowest
- Recycling → **Φ** effect on **PBT** and **PLA, decrease** for **PET** and **PBS (**degradation+↑crystalline)
- 1X extr MMT → **decrease** in all samples, especially for **PBS** because of the agglomerates.
- 2X extr MMT → **decrease** for **PET** and **PLA.**
  → **increase** for **PBT** and **PBS** , Improved distribution+ size of aggregates

Practical part

Flexural strength and flexural modulus of composites (PET, PBS, PBT, PLA / MMT)





- 1X extr MMT → decrease in all samples except for **PBS**, which had no effect.
- 2X extr & 2X extr MMT: **Φ** effect the **flexural strength** of polyesters and their nanocomposites.
- **PLA/MMT** is the stiffest while **PBS** is the most flexible
- Reprocessing did not lead to a significant change in the **flexural modulus** of polyester.
- 1X extr MMT → **increase** in the stiffness of all polyesters**,** increased crystallinity**.**
- 2X extr MMT → **increase** for **PET**, **PBS** and **PBT.**
  → **decrease** for **PLA.**

Scanning Electron Microscope (SEM) - Energy Dispersive Spectroscopy (EDS) analysis of composites (PET, PBS, PBT, PLA / MMT)

Practical part

PET

Flat surface

PLA

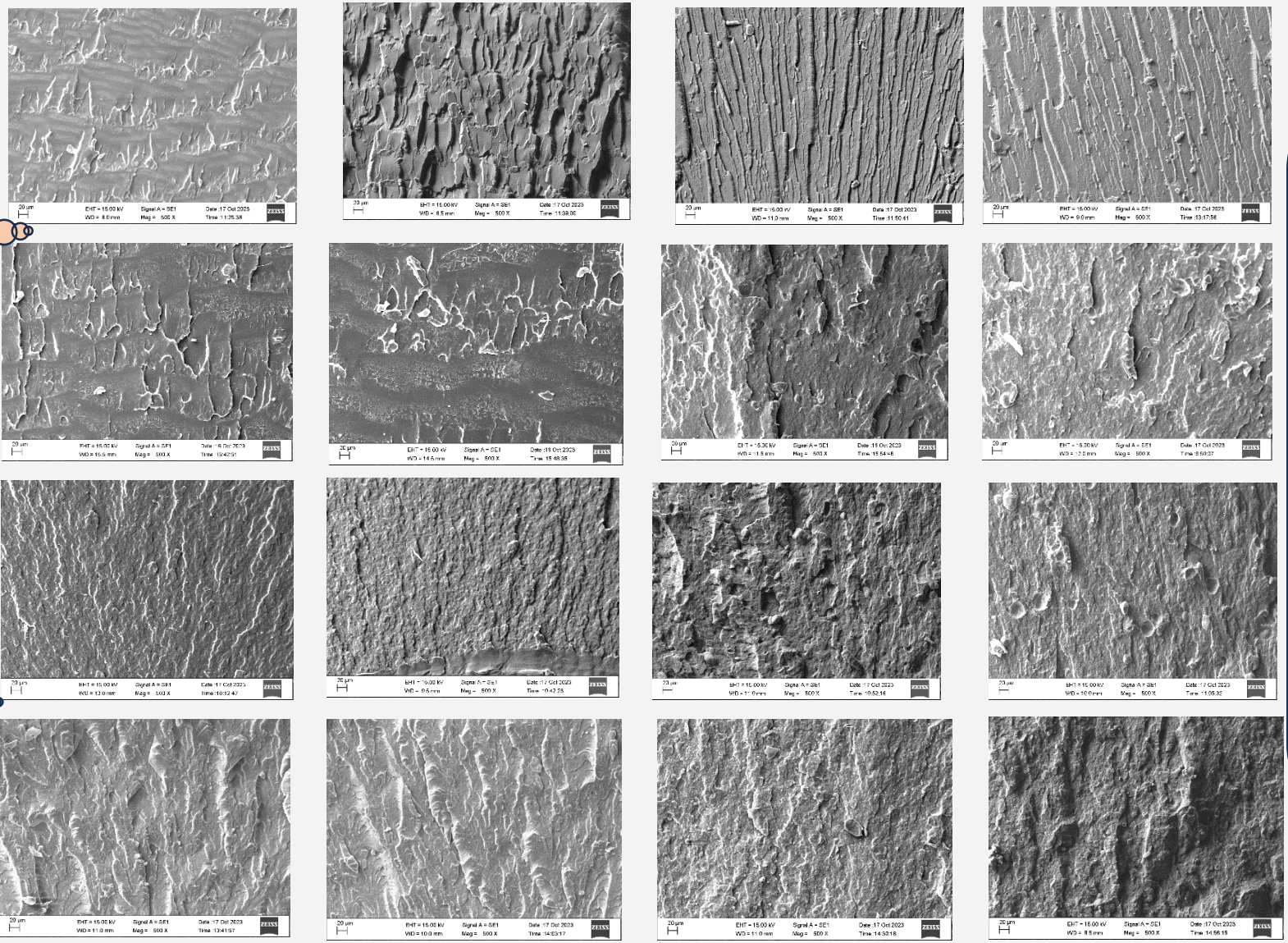
Izod impact

PBS

Rough surface

PBT

1X extr | **2X extr** | **1X extr MMT** | **2X extr MMT**

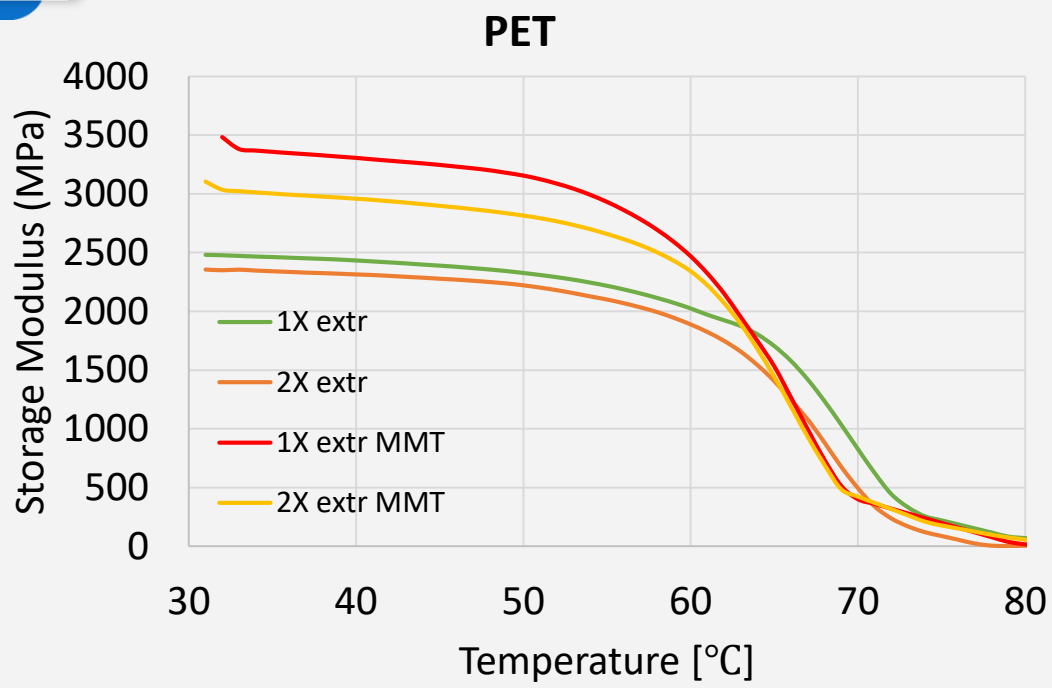


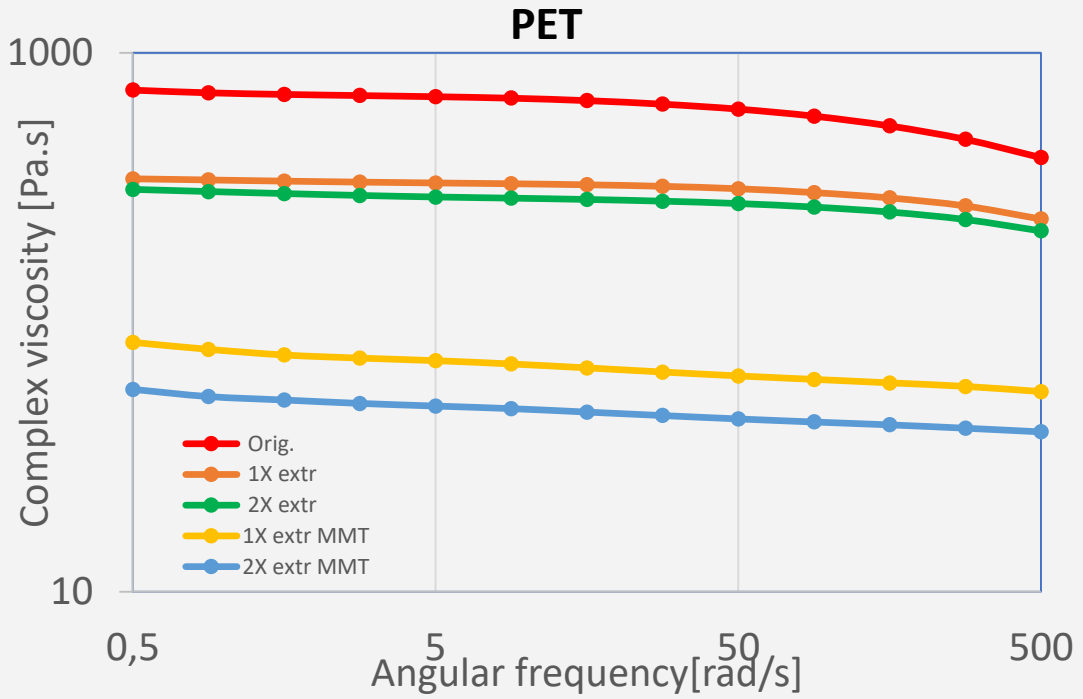
- **2X extr** → the fracture surfaces **did not change** significantly, except for **PET** (brittle)
- **1X extr MMT** → **reduced** plastic deformation.
- **2X extr MMT** → the fracture surfaces of the nanocomposites **did not change.**

Practical part

Time- and temperature-dependent properties of MMT/polyester nanocomposites

I measured the storage modulus and complex viscosity of polyesters and polyesters/MMT using DMA and Rheometer.





- Reprocessing and the presence of MMT have an effect on the storage modulus and complex viscosity.

The results

Conclusion:

- The results indicate a change in the properties of polyesters as a result of **the following factors:** Adding MMT and recycling.
- Recycling → change in the distribution of MMT in the polyester matrix → change in the final properties of nanocomposites.
- The difference in the effect of MMT and recycling depends on the type of polyester used.
- MMT had a nucleation effect so the crystallization temperature of all composites increased when it was added.
- **PBS** had higher crystallinity, more flexibility, higher impact strength and rougher fracture surface compared to other polyester materials used.
- The polyester that tends to increase its chain order is **PET** due to its low viscosity
- The presence of MMT in the polyester matrix changed the properties of the nanocomposites after reprocessing, increasing them in some cases to compensate for molecular degradation such as impact strength **PBS/MMT** and **PBT/MMT**.
- Adding MMT to polyesters increased the stiffness, especially **PLA**.

Plans for the future

I will do the following

- Further analyses of new results.

- Scan the samples by using Wide-angle X-ray scattering (WAXD).

- Studying the effect of recycling and adding MMT on the rheology of samples.

- Write two articles

1- Effect of reprocessing on the viscosity and mechanical properties of PLA and PLA/MMT nanocomposites.

2- Recycling of petroleum- and bio-based polyester / montmorillonite nanocomposites.

- start writing the thesis.

List of publication

List of publication

- **First article :** *Zoubeida Taha Taha, Andrea Ádámné Major*, **A review on MWCNTs: The effect of its addition on the polymer matrix,** *Gradus*, https://gradus.kefo.hu/archive/2023-1/2023_1_ENG_012_Taha.pdf

- **Second article :** *Zoubeida Taha Taha, Andrea Ádámné Major*, **Investigating the effect of adding multiwalled carbon nanotubes on the morphological properties of polybutylene terephthalate,** Springer, Advanced Sciences and Technologies for Security Applications, Critical Infrastructure Protection in the Light of the Armed Conflicts, https://link.springer.com/chapter/10.1007/978-3-031-47990-8_41.

- **Third article :** *Zoubeida Taha Taha, Andrea Ádámné Major, Ferenc Ronkay,* **Effect of Reprocessing on the Crystallization of Different Polyesters,** *Acta Technica Jaurinensis,* https://acta.sze.hu/index.php/acta/article/view/723/620 .

- **Fourth article:** *Zoubeida Taha Taha, Attila Bata, Béla Molnár, Ferenc Ronkay,* ***Comparing petroleum- and bio-based polyesters and their montmorillonite nanocomposites for recyclability****, Cleaner and Environmental Systems,* **(under review).**

Semester Activities

Semester Activities

- I explored the literature review related to my research topic and wrote summaries of it.
- I wrote an article on my research results and started the second one.
- I continued to carry out experiments and measurements.
- I evaluated the results.

THANK YOU for YOUR ATTENTION!