Open Rubric

**Table 6.4.** Tensile and impact data of neat PHBH, binary and hybrid composites

| Sample | Tensile modulus /MPa | Elongation at beak /% | Impact resilience /kJ.m$^{-2}$ |
|---|---|---|---|
| PHBH | 748.2 ± 49.4 | 7.6 ± 0.6 | 5.2 ± 0.6 |
| PHBH/SB | 1063.8 ± 18.5 | 4.7 ± 0.3 | 4.5 ± 0.4 |
| PHBH/SD | 1071± 113 | 5.1 ± 0.5 | 4.5 ± 0.5 |
| PHBH/SB/SD | 945 ± 84.5 | 4.2 ± 0.6 | 4.4 ± 0.2 |
| PHBH/SB/SD/HS | 914.5 ± 42.1 | 4.5 ± 0.2 | 4.4 ± 0.3 |

## 6.4. Conclusions

The investigation of the morphology showed a high interfacial tension in the hybrid composites due to the hydrophobicity of PHBH and the hydrophilic nature of MSF and SB, but the PHBH/SD composite showed better dispersion of the filler. The differences in morphologies account for the variations in composite performance. The presence of HS in the hybrid composite (PHBH/SB/SD/HS) promoted interfacial adhesion between PHBH and SD. Compared to pristine PHBH, the binary and hybrid composites demonstrated superior stiffness, viscosity and storage modulus. The thermal stability of PHBH was enhanced with the addition of SB and SD, but the mixture of the fillers showed a synergistic effect on thermal stability. Contrarily, the presence of HS in the PHBH/SB/SD/HS achieved higher thermal stability of the composite due to the barrier effect of HS. The addition of fillers resulted in a reduction in elongation at break and impact resilience of PHBH, although the elongation of the composites remained within a similar range. The reduction in mechanical properties provides evidence of high interfacial tension between PHBH and either SB or SD in the hybrid composites. In future work, maleic anhydride grafted poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBH-g-MA) will be produced to investigate its effectiveness as a compatibiliser at various loadings for PHBV/SB, PHBH/SD, PHBH/SB/SD, and PHBH/SB/SD/HS composites prepared through melt extrusion.

## 6.5. References

Elfaleh, I., Abbassi, F., Habibi, M., Ahmad, F., Guedri, M., Nasri, M. & Garnier, C. **2023**. A comprehensive review of natural fibers and their composites: An eco-friendly alternative to conventional materials. *Results in Engineering*, 19(101271): 101271–101271. DOI: 10.1016/j.rineng.2023.101271.

Empty-Marin, D., Chiarello, L. M., Wiggers, V. R., Dantas, A. & Botton, V. **2023**. Effect of coupling agents on properties of vegetable fiber polymeric composites: review. *Polimeros-ciencia E Tecnologia*, 33(1).

Fu, S. -Y., Lauke, B., Mai, Y. -W. & Yue, C. -Y. **2009**. Science and engineering of short fibre-reinforced polymer composites. Oxford: Woodhead Publishing. DOI: 10.1533/9781845696498.1.

Girijappa Thyavihalli, Y. G., Mavinkere Rangappa, S., Parameswaranpillai, J. & Siengchin, S. **2019**. Natural fibers as sustainable and renewable resource for development of eco-friendly composites: A comprehensive review. *Frontiers in Materials*, 6(266): 1–14. DOI: 10.3389/fmats.2019.00226.

Hilliou, L. & Covas, J. A. **2020**. In-process rheological monitoring of extrusion-based polymer processes. *Polymer International*, 70(1): 24–33. DOI: 10.1002/pi.6093. **5**

Hu, Y., Zhang, J. M., Sato, H., Noda, I. & Ozaki, Y. **2007**. Multiple melting behavior of poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) investigated by differential scanning calorimetry and infrared spectroscopy. *Polymer*, 48 (16): 4777–4785. DOI: 10.1016/j.polymer.2007.06.016.

Ivorra-Martinez, J., Manuel-Mañogil, J., Boronat, T., Sanchez-Nacher, L., Balart, R. & Quiles-Carrillo, L. **2020**. Development and characterization of sustainable composites from bacterial polyester poly(3-Hydroxybutyrate-co-3-hydroxyhexanoate) and almond shell flour by reactive extrusion with oligomers of lactic acid. *Polymers,* 12(5): 1097. DOI: 10.3390/polym12051097.

Kun, D., Kárpáti, Z., Fekete, E. & Móczó, J. **2021**. The role of interfacial adhesion in polymer composites engineered from lignocellulosic agricultural waste. *Polymers*, 13(18): 3099. DOI: 10.3390/polym13183099.

Liu, K., Lai, S., Han, J. & Hsieh, K. **2020**. Properties of sugarcane fiber/polyurethane-crosslinked epoxy composites under different interfacial treatments. *Polymer composites*, 41(10): 4277–4287. DOI: 10.1002/pc.25710.

Mariana, M., Alfatah, T., Abdul Khalil, H. P. S., Yahya, E. B., Olaiya, N. G., Nuryawan, A., Mistar, E. M., Abdullah, C. K., Abdulmadjid, S. N. & Ismail, H. **2021**. A current advancement on the role of lignin as sustainable reinforcement material in biopolymeric blends. *Journal of Materials Research and Technology*, 15: 2287–2316. DOI: 10.1016/j.jmrt.2021.08.139.

Masanabo, M. A., Tribot, A., Luoma, E., Virkajärvi, J., Sharmin, N., Sivertsvik, M., Ray, S. S., Keränen, J. & Emmambux, M. N. **2024.** Development and characterization of poly(butylene succinate-co-adipate)/poly(3-hydroxybutyrate-co-3-hydroxyvalerate) with cowpea lignocellulosic fibers as a filler via injection molding and extrusion film-casting. *Macromolecular Materials and Engineering*, 309 (8): 240037. DOI: 10.1002/mame.202400037.

Medina-Martinez, C. J., Sandoval Herazo, L. C., Zamora-Castro, S. A., Vivar-Ocampo, R. & Reyes-Gonzalez, D. **2023**. Use of sawdust fibers for soil reinforcement: A review. *Fibers*, 11(7): 58. DOI: 10.3390/fib11070058.

Mochane, M. J., Mokhena, T. C., Mokhothu, T. H., Mtibe, A., Sadiku, E. R., Ray, S. S., Ibrahim, I. D. & Daramola, O. O. **2019**. Recent progress on natural fiber hybrid composites for advanced applications: A review. *Express Polymer Letters*, 13(2): 159–198. DOI: 10.3144/expresspolymlett.2019.15.

Mohlala, L. M., Bodunrin, M. O., Awosusi, A. A., Daramola, M. O., Cele, N. P., & Olubambi, P. A. **2016**. Benefifciation of corncob and sugarcane bagasse for energy generation and materials development in Nigeria and South Africa: A short overview. *Alexandria Engineering Journal*, 55(3): 3025–3036. DOI: 10.1016/j.aej.2016.05.014.

Musthaq, M. A., Dhakal, H. N., Zhang, Z., Barouni, A. & Zahari, R. **2023**. The effect of various environmental conditions on the impact damage behaviour of natural-fibre-reinforced composites (NFRCs)—A critical review. *Polymers*, 15(5): 1229. DOI: 10.3390/polym15051229.

Patel, R. V., Yadav, A. & Winczek, J. **2023**. Physical, mechanical, and thermal properties of natural fiber-reinforced epoxy composites for construction and automotive applications. *Applied Sciences*, 13(8): 5126. DO: 10.3390/app13085126.

Pawlak, A. & Krajenta, J. **2024**. Entanglements of macromolecules and their influence on rheological and mechanical properties of polymers. *Molecules*, 29(14): 3410–3410. DOI: 10.3390/molecules29143410.

Pokharel, A., Falua, K. J., Babaei-Ghazvini, A. & Acharya B. **2022**. Biobased polymer composites: A review. *Journal of Composites Science*, 6(9): 255. DOI: 10.3390/jcs6090255.

Samanth, M., Hiremath, P., Divya Deepak, G., Naik, N., Arunkumar, S. H., Heckadka, S. S. & Shivamurthy, R. C. **2025**. Sustainable composites from sugarcane bagasse fibers and bio-based epoxy with insights into wear performance, thermal stability, and machine learning predictive modeling. *Journal of Composites Science*, 9(3): 124–124. DOI: 10.3390/jcs9030124.

Seydibeyoğlu, M. Ö., Dogru, A., Wang, J., Rencheck, M., Han, Y., Wang, L., Seydibeyoğlu, E. A. A., Zhao, X., Ong, K., Shatkin, J., Es-haghi, S. S., Bhandari, S., Ozcan, S. & Gardner, D. J. **2023**. Review on hybrid reinforced polymer matrix composites with nanocellulose, nanomaterials, and their fibers. *Polymers* 15(4): 984–984. DOI: 10.3390/polym15040984.

Shamsuyeva, M., Hansen, O. & Endres, H. -J. Review on hybrid carbon/flax composites and their properties. **2019**. *International Journal of Polymer Science*, (2019): 1–17. DOI: 10.1155/2019/9624670.

Shelly, D., Lee, S. & Park, S. **2025**. Hemp fiber and its bio-composites: a comprehensive review part I—Characteristics and processing. *Advanced Composites and Hybrid Materials*, 8(3). DOI: 10.1007/s42114-025-01314-0.

Sikhosana, T. S., Malebo, N. J., Motloung, M. P., Mofokeng, T. G. & Mochane, M.J. **2025**. The influence of halloysite clay on the properties of the polybutylene succinate (PBS)/sawdust, PBS/sugarcane bagasse, and PBS/sawdust/sugarcane bagasse hybrid composites. *Polymers*, 17(15): 2120. DOI: 10.3390/polym17152120.

Theys, L., Mochane, M. J., Mofokeng, T. G., Motloung, M. T., Motloung, M. P. & Mokhena, T. C. **2025**. The effect of expandable graphite and montmorillonite (MMT) clay on the morphology, thermal stability, and flammability properties of the maize stalk/PBS bio-composite. *Journal of Thermoplastic Composite Materials*, 0(0). DOI: 10.1177/08927057251329245.

Valente, B. F. A., Silvestre, A. J. D., Neto, C. P., Vilela, C., & Freire, C. S. R. **2021**. Effect of the Micronization of Pulp Fibers on the Properties of Green Composites. *Molecules*, 26 (18): 5594. DOI: 10.3390/molecules26185594.

Yang, Y., Chen, Y., Leng, F., Huang, L., Wang, Z. & Tian, W. **2017**. Recent advances on surface modification of halloysite nanotubes for multifunctional applications. *Applied Sciences*, 7(12): 1215. DOI: 10.3390/app7121215.

# CHAPTER 7

## Flammability properties of the PHBH and PBS systems

It is well known that fire is a catalyst that thermally oxidises polymer-based materials. Flame-retardant materials are capable of withstanding high temperatures and resist burning. Cone calorimeter is one of the techniques used to analyse the flammability properties of the polymer-based composites. A lower peak heat release rate value is an indication of a flame-retardant system, while a higher peak heat release rate value is an indication of a flammable material (Araby *et al*., 2021). The well-known parameters from the cone calorimeter include heat release rate (HRR), total heat release (THR), mass loss rate (MLR), and smoke production rate (SPR). **Table 1** summarises the cone calorimeter parameters for the PBS based system. Both natural fiber-reinforced composites, specifically sugarcane bagasse (SB) and sawdust (SD), demonstrated higher peak heat release rate values (**Table 1**) when compared to neat poly(butylene succinate) (PBS). This increase is primarily due to the inherent flammability of lignocellulosic materials, which decompose rapidly and act as catalysts during combustion, accelerating the burning process (Mankeed *et al*., 2022). When combined, SB and SD exhibited even greater flammability, likely resulting from the synergistic degradation of their similar organic components (Alias *et al*., 2021; Mankeed *et al*., 2022).

**Table 7.1.** Selective cone calorimeter parameters for PBS based composites

| **Samples ID** | **pHRR / kW.m$^{-2}$** | **TTI /s** | **Total heat release /MJ.m$^{-2}$** | **FPI /m$^2$s.k$^{-1}$W$^{-1}$** |
|---|---|---|---|---|
| PBS | 468 | 64 | 99.9 | 0.1368 |
| PBS/SB | 578.2 | 62 | 111.4 | 0.1072 |
| PBS/SD | 505.8 | 57 | 92.8 | 0.1127 |
| PBS/HS | 425.3 | 95 | 78.9 | 0.2234 |
| PBS/SB/SD | 576.3 | 62 | 111.6 | 0.1076 |
| PBS/SB/HS | 451.9 | 67 | 134.2 | 0.1483 |
| PBS/SD/HS | 432.8 | 65 | 92.1 | 0.1438 |
| PBS/SB/SD/HS | 436.1 | 71 | 90.1 | 0.1628 |
| PBS/SB/SD/HS/EG | 370 | 89 | 93.3 | 0.2405 |

Nevertheless, the incorporation of halloysite nanotubes (HS) into both binary and hybrid composite systems led to a significant reduction in pHRR, indicating improved flame retardancy. Halloysite nanotubes, in particular, have been shown to enhance the thermal stability of polymer matrices due to their tubular structure and high aspect ratio, which improve heat barrier properties (Kumar *et al*., 2024; Marset *et al*., 2020). Further enhancements in flame retardancy were noted with the incorporation of expandable graphite (EG). The addition of expandable graphite contributes to intumescent action; upon heating, EG expands to form a foamed, carbonaceous char that insulates the underlying polymer (Kmeťová *et al*., 2025). Among all samples examined, the hybrid composite comprising PBS/SB/SD/HS/expandable graphite (EG) showed the lowest pHRR value of approximately 370 $kW \cdot m^{-2}$, highlighting enhanced fire resistance. In addition to the reduced heat release, this hybrid system also demonstrated a relatively high Fire Performance Index (FPI) value of 0.2405 $m^2 \cdot s \cdot kW^{-1}$, which indicates improved flame retardancy. The FPI, defined as the time to ignition divided by the peak heat release rate (pHRR), determines material safety performance. Generally, higher FPI values are associated with delayed ignition and a slower heat release, indicating a lower fire hazard (Murad *et al*., 2025). The exceptional flame retardancy observed in the PBS/SB/SD/HS/EG composite is attributed to the formation of a stable, compact, and continuous carbonised ceramic char layer. This char residue serves as an effective thermal and physical barrier, inhibiting heat and mass transfer between the flame and the underlying material. It also traps volatile degradation products, reducing the availability of flammable gases during combustion. This combined effect yields a significant improvement in flame-retardant performance across multiple key parameters. Another crucial metric alongside the peak heat release rate (pHRR) measurement in the fire performance of composites is the total heat release (THR). In the absence of flame retardants, the composites showed high THR values, with the sugarcane bagasse (SB)-reinforced composites exhibiting notably higher THR compared to those containing sawdust (SD) (**Table 1**). This difference can be attributed to the unique chemical compositions and thermal degradation behaviours of the lignocellulosic fillers (Mankeed *et al*., 2022). Sugarcane bagasse usually contains a higher proportion of hemicellulose, and a lower mineral content compared to sawdust (Murad *et al*., 2025). As a result, it undergoes more extensive thermal degradation and releases more combustible volatiles during combustion (Becker *et al*., 2010). The inclusion of flame retardants significantly reduced the THR values across the composites. Notably, the PBS/SB/SD/HS/EG blend showed the most pronounced reduction in THR. This flame-retardant system enhances thermal stability and reduces heat release through multiple mechanisms. Halloysite nanotubes contribute to thermal stability by reinforcing the char structure and potentially facilitating the formation of a cohesive, ceramic-like residue during combustion (Marset *et al*., 2020). Expandable graphite serves as an intumescent agent, expanding upon heating to create an insulating char layer that shields the underlying material from flames (Kmeťová *et al*., 2024).

Together, these additives inhibit combustion and reduce the production of flammable decomposition products, thereby lowering total heat release (THR) and improving flame resistance. One of the most common hazardous gases released during combustion is carbon monoxide (CO), primarily because it binds to haemoglobin in the blood, reducing oxygen delivery to tissues. Carbon monoxide poisoning can lead to serious consequences, including severe tissue damage, organ failure, and even death, and therefore measurements are necessary (Afzal *et al*., 2025). **Figure 1** depicts carbon monoxide production (COP) curves. The first two peaks correspond to initial pyrolysis and flaming combustion, where incomplete combustion results in high carbon monoxide (CO) levels. The third peak occurs during smouldering, with limited oxygen causing inefficient combustion and a temporary drop in CO as it converts to carbon dioxide ($CO_2$) (Wyn *et al*., 2020). The PBS/SB/SD/HS/EG composite system produced the least CO due to the formation of a protective, ceramic-like char layer. The addition of expandable graphite (EG) and hybrid synergists further enhances this flame-retardant effect.

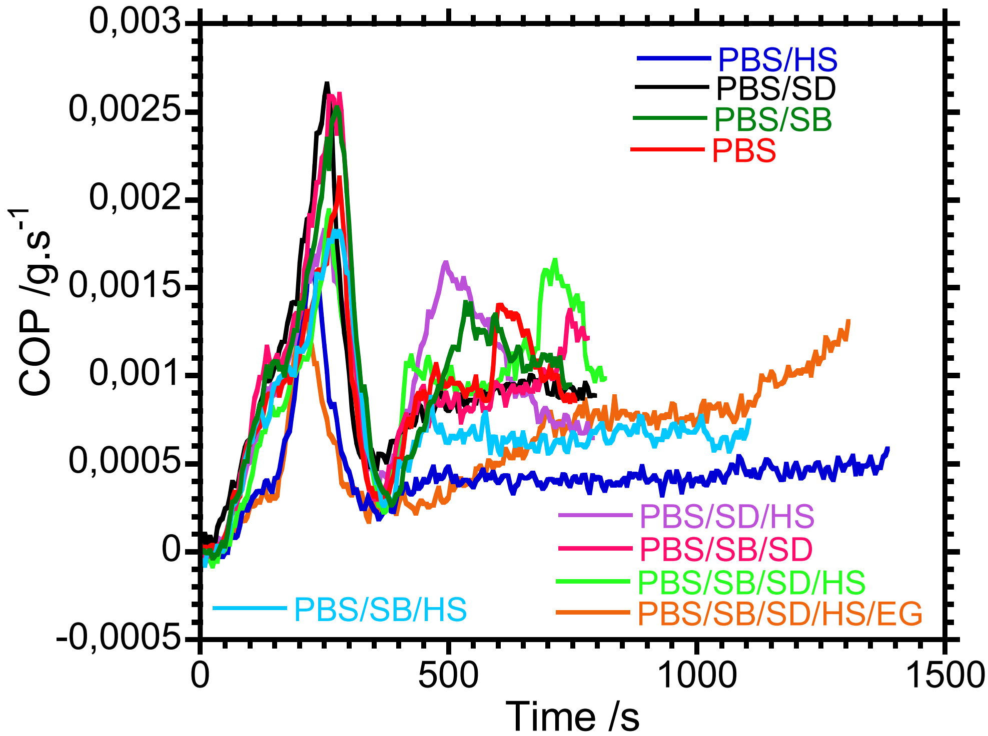


**Figure 7.1.** Carbon monoxide production of the investigated samples

**Table 2** depicts the selective cone calorimeter parameters for PHBH-based system. The addition of single fibres, i.e., sugarcane bagasse (SB) and sawdust (SD), to the poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBH) matrix, resulted in higher pHHR values compared to the neat polymer, which was similar to the PBS-based system. This increase may be related to the high cellulose and hemicellulose content of lignocellulosic fibres, which readily decompose under heat during pyrolysis, releasing more combustible volatiles and intensifying the combustion process (Mankeed *et al*., 2022; Nurazzi *et al*., 2021), as explained elsewhere in this document.

**Table 7.2.** Selective cone calorimeter parameters for PHBH based system

| **Samples ID** | **pHRR / $kW.m^{-2}$** | **TTI /s** | **Total heat release /$MJ.m^{-2}$** | **FPI /$m^2s.k^{-1}W^{-1}$** |
|---|---|---|---|---|
| PHBH | 508.1 | 55 | 85.15 | 0.108 |
| PHBH/SB | 564.1 | 43 | 115 | 0.076 |
| PHBH/SD | 571.4 | 42 | 116.2 | 0.073 |
| PHBH/SB/SD | 545.1 | 48 | 110.9 | 0.088 |
| PHBH/SB/SD/HS | 509.7 | 46 | 111.1 | 0.090 |

When fibres were combined in the PHBH matrix (PHBH/SB/SD), the peak heat release rate (pHRR) slightly decreased compared to single-fibre composites, which contradicts the findings in the PBS-based system. This phenomenon indicates that the synergistic interactions between the two fibre types could affect the degradation pathway, thereby limiting heat and mass transfer during combustion (Kumar *et al*., 2024). One may suggest that there is a better interaction of fibres in this system, which might have blocked any volatile materials from leaving the system and, in the process, slightly enhanced the flame retardancy. Further improvement in flame retardancy was observed with the addition of halloysite nanotubes (HS) to the fibre-reinforced PHBH system. The PHBH/SB/SD/HS composite exhibited the lowest pHRR among all the composites studied, although its value remained slightly higher than that of the neat PHBH.

This indicates that while halloysite clay contributes to improved thermal stability through its barrier effect, the combined flammability of the natural fibres still plays a significant role in influencing the overall fire behaviour of the composite (Nurazzi *et al*., 2021). Halloysite clay enhances flame retardancy by acting as a heat barrier and promoting char formation, but its effectiveness can be limited by the abundance of easily combustible organic fibre content (Kumar *et al*., 2024; Marset *et al*., 2020). Notably, the Fire Performance Index (FPI) of the PHBH/SB/SD/HS system was lower than that of the neat polymer, indicating a higher fire risk despite the reduced relative risk (RR). FPI is defined as the ratio of time to ignition (TTI) to pHRR and is commonly used to assess the fire growth potential of materials. A lower FPI value indicates faster ignition and/or higher heat release, both of which contribute to rapid fire development (Huang *et al*., 2023). This observation suggests that although the composite burns with lower intensity (lower pHRR), it ignites more quickly or lacks sufficient delay in ignition, likely due to the volatile degradation products of the fibres. Overall, while the addition of halloysite improves the thermal barrier properties and reduces the pHRR, the reduced FPI implies that additional flame-retardant strategies may be necessary. Combining halloysite with intumescent agents, such as expandable graphite, could be an effective approach to delaying ignition and suppressing the early stages of fire growth.

## Conclusions

The flammability analysis revealed significant performance variations between the PBS and PHBH composite systems. In both matrices, the addition of natural fibres increased the peak heat release rate (pHRR) due to the lignocellulosic fillers' combustibility. However, halloysite nanotubes (HS) consistently enhanced flame retardancy by promoting char formation and acting as a thermal barrier. The benefits of flame-retardant synergy were greater for PBS. For instance, HS alone reduced the pHRR by approximately 9%. In the PBS/SB/SD/HS/EG system, there was a 21% reduction in pHRR and a 76% increase in the Fire Performance Index (FPI). Conversely, the PHBH composites (evaluated without EG) exhibited only moderate improvements. In this case, HS provided about a 6% reduction in pHRR compared to the fibre-reinforced control. This comparison highlights that while halloysite alone can enhance thermal stability, its combination with an intumescent additive is crucial for achieving high fire resistance and maintaining balanced flammability performance. These findings have direct implications for fire-sensitive applications. PBS-based hybrids containing HS and EG show promise for use in packaging, construction panels, and automotive components, where reduced heat release and improved char integrity can help slow fire growth and enhance safety in real-world conditions.

## References

Afzal, M., Agarwal, S., Elshaikh, R.H., Babker, A. M. A., Choudhary, R. K., Prabhakar, P. K., Zahir, F. & Sah, A. K. **2025**. Carbon monoxide poisoning: Diagnosis, prognostic factors, treatment strategies, and future perspectives. *Diagnostics*, 15(5): 581. DOI: 10.3390/diagnostics15050581.

Alias, A. H., Norizan, M. N., Sabaruddin, F. A., Asyraf, M. R. M., Norrahim, M. N. F., Ilyas, A. R., Kuzmin, A. M., Rayung, M., Shazleen, S. S., Nazrin, A., Sherwani, S. F. K., Harussani, M. M., Atikah, M. S. N., Ishak, M. R., Sapuan, S. M. & Khalina, A. **2021**. Hybridization of MMT/Lignocellulosic fiber reinforced polymer nanocomposites for structural applications: A review. *Coatings*, 11(11): 1355. DOI: 10.3390/coatings11111355.

Araby, S., Philips, B., Meng, Q., Ma, J., Laoui, T. & Wang, C. **2021**. Recent advances in carbon-based nanomaterials for flame retardant polymers and composites. 212: 108675–108675. DOI: 10.1016/j.compositesb.2021.108675.

Becker, C., Sharma, L. N. & Chambliss, C. K. **2010**. Analytical monitoring of pretreatment and hydrolysis processes in lignocellulose-to-bioalcohol production. Elsevier eBooks, pp.281–314. DOI: 10.1533/9781845699611.4.281.

Huang, Q., Li, X., Han, P., Li, Y., Liu, C., Chen, Q. & Li, Q. **2023**. Research on the fire behaviors of polymeric separator materials PI, PPESK, and PVDF. *Fire*, 6(10): 386. DOI: 10.3390/fire6100386.

Kumar, I., Tirlangi, S., Kathiresan, K., Sharma, V., Madhu, P., Sathish, T., Ağbulut, Ü. & Murugan, P. **2024**. Co-pyrolysis of furniture wood with mixed plastics and waste tyres: assessment of synergistic effect on biofuel yield and product characterization under different blend ratio. *Scientific Reports*, 14(1). DOI: 10.1038/s41598-024-72809-x.

Kmeťová, E., Kačíková, D. & Kačík, F. **2024**. The effect of intumescent coating containing expandable graphite onto spruce wood. *Coatings*, 14(4): 490–490. DOI: 10.3390/coatings14040490.

Mankeed, P., Onsree, T., Naqvi, S.R., Shimpalee, S. & Tippayawong, N. **2022**. Kinetic and thermodynamic analyses for pyrolysis of hemp hurds using discrete distributed activation energy model. *Case Studies in Thermal Engineering*, 01870. DOI: 10.1016/j.csite.2022.101870.

Marset, D., Dolza, C., Fages, E., Gonga, E., Gutiérrez, O., Gomez-Caturla, J., Ivorra-Martínez, J., Sànchez-Nácher, L. & Quiles-Carrillo, L. **2020**. The effect of halloysite nanotubes on the fire retardancy properties of partially biobased polyamide 610. *Polymers*, 12(12): 3050–3050. DOI: 10.3390/polym12123050.

Murad, M. S., Hamzat, A. K., Asmatulu, E. & Asmatulu, R. **2025**. Flame-retardant fiber composites: synergistic effects of additives on mechanical, thermal, chemical, and structural properties. *Advanced Composites and Hybrid Materials*, 8(1). DOI: 10.1007/s42114-024-01111-1.

Nurazzi, N. M., Asyraf, M. R. M., Rayung, M., Norrrahim, M. N. F., Shazleen, S. S., Rani, M. S. A., Shafi, A. R., Aisyah, H. A., Radzi, M. H. M., Sabaruddin, F. A., Ilyas, R. A., Zainudin, E. S. & Abdan, K. **2021**. Thermogravimetric analysis properties of cellulosic natural fiber polymer composites: A review on influence of chemical treatments. *Polymers*, 13(16): 2710. DOI: 10.3390/polym13162710.

Wyn, H. K., Konarova, M., Beltramini, J., Perkins, G. & Yermán, L. **2020**. Self-sustaining smouldering combustion of waste: A review on applications, key parameters and potential resource recovery. *Fuel Processing Technology*, 205: 106425. DOI: 10.1016/j.fuproc.2020.106425.

# CHAPTER 8

## Conclusions and future recommendations

The study investigated the development of sustainable, flame-retardant, natural fibre-reinforced composites using agricultural residues and inorganic flame-retardant additives. The primary objective was to assess the effects of various flame-retardant fillers, expandable graphite (EG), and halloysite nanotubes (HS) on the flammability, structural integrity, and thermal performance of bio-based composites composed of polybutylene succinate (PBS) and poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBH), which were reinforced with sugarcane bagasse (SB) and sawdust (SD). A range of techniques, including FTIR, SEM, XRD, TGA, rheology, mechanical testing, and cone calorimetry, were employed to assess thermal, structural, and interfacial behaviours. In PBS composites, SD and SB enhanced stiffness and viscosity, with EG and HS contributing to crystallisation and interfacial strength. Hybridising the fibres resulted in improved filler dispersion and melt strength, though weak fibre–matrix adhesion limited toughness. EG increased crystallisation rates, aiding processing, while HS improved structural integrity. Despite slight reductions in thermal stability due to filler degradation, PBS hybrids demonstrated promising properties for rigid applications. In PHBH composites, SD again showed better compatibility than SB, while HS contributed to improved cohesion and thermal stability. Although hybrid systems enhanced stiffness and processing stability, their mechanical performance was limited by brittleness and poor interfacial bonding.

These findings highlight the need for coupling agents, such as PHBH-g-MA, to improve fibre-matrix integration. In conclusion, the analysis revealed that SD exhibited greater compatibility across both matrices, whereas SB contributed to improved mechanical stiffness. Hybrid systems outperformed their single-fibre counterparts, although performance varied according to the matrix utilised. PBS enabled more effective fibre integration, while PHBH necessitated more precise interfacial adjustments. Furthermore, the incorporation of flame retardants into fibre–polymer systems effectively suppressed combustion, as evidenced by the reduced peak heat release rate (pHRR). Although halloysite nanotubes demonstrated heat barrier-forming capabilities, their synergy with intumescent additives, such as expandable graphite, proved to be more effective and promising. This research demonstrates that, with appropriate selection of fillers and processing strategies, agricultural waste can be repurposed into biodegradable, flame-retardant composites that are well-suited for sustainable engineering applications such as packaging, construction, and interior applications.

Future work could further investigate industrial-scale processing, specifically the optimisation of extrusion and injection moulding. This research will help validate reproducibility and cost-effectiveness under real manufacturing conditions. Additionally, long-term durability and environmental ageing studies are essential. These should include hydrothermal, UV, and biodegradation testing to assess material stability and performance throughout its service life. These efforts will aid in transitioning the developed composites from laboratory-scale formulations to commercially viable and sustainable material alternatives.