

**The doctoral dissertation of M.Sc. Eng. Kacper Fiedurek titled:
Development of the Formulation and Processing Technology for Flame-
Retardant, Machinable Biodegradable Polymer Composites for
Engineering Applications**

ABSTRACT

This doctoral dissertation concerned the development of a formulation and a technological basis for producing a composite based on biodegradable polylactide (PLA) with reduced flammability and suitability for machining. The research addressed the need to extend the engineering applications of PLA, which are limited by its flammability, sensitivity to processing conditions and local softening during machining. The aim was to reduce combustion intensity while retaining properties relevant to the manufacture and subsequent shaping of functional components.

The literature review indicated that research on flame-retardant PLA focuses primarily on additive selection and the assessment of combustion parameters. Less attention has been given to the relationships between raw material preparation, extrusion conditions, composite structure and machining behaviour. The research gap therefore concerned the combined consideration of material formulation and processing technology as factors determining end-use properties.

The central hypothesis was that appropriate formulation and control of extrusion parameters could yield modified PLA with reduced flammability and suitability for machining. The scientific objective was to determine how graphite and phosphorus-based additives, the physical form of the feedstock and process settings influence composite structure and properties. The practical objective was to establish a basis for its manufacture and identify processing conditions that balance material properties against process loads.

Preliminary studies compared graphite and phosphorus-based additives, assessed the suitability of single-screw and twin-screw extrusion, and examined the importance of grinding PLA before blending the constituents. Process observations and scanning electron microscopy (SEM) images showed that using a powdered matrix reduced mixture segregation and promoted a more uniform distribution of additives. These findings supported the selection of co-rotating twin-screw extrusion. Based on cone calorimetry, a composite containing 80 wt% PLA, 10 wt% E90 expandable graphite and 10 wt% CP organic phosphorus flame retardant (AFLAMMIT

PCO 962) was selected for the main study. This formulation reduced heat release more effectively than the previously investigated composition containing GrafGuard graphite and ammonium polyphosphate.

The main study followed a three-factor Hartley design comprising 11 extrusion runs. Feed rate, screw speed and temperature setpoint were varied while the composite formulation remained constant. Drive loading, melt temperature, melt mass-flow rate (MFR), and tensile and flexural properties were found to depend on the processing conditions. MFR ranged from 28.35 to 65.43 g/10 min. Compared with unmodified PLA, peak heat release rate decreased by approximately 63–72%, total heat release by 13–47%, and mass loss rate by 59–68%. The reduction in combustion intensity was, however, accompanied by earlier ignition and increased smoke production.

Regression models were developed and compared to describe the effects of extrusion settings on the responses under study, and their predictive performance was assessed using leave-one-out cross-validation (LOO). An excellent fit of a full second-order model did not always correspond to the lowest prediction error. Multi-criteria selection of process settings, based on desirability functions for eleven responses, revealed that the individual optima did not coincide. With equal criterion weights, the full models identified a compromise on the adopted grid at a feed rate of 3.5 kg/h, a screw speed of 175 rpm and a temperature setpoint of 173°C. The result depended on model form, and the selected settings require experimental confirmation.

Drilling and milling trials complemented the assessment of the composite's suitability for further shaping. Under the conditions studied, the composite specimens exhibited more favourable material separation and less extensive residual material at the edges than unmodified PLA specimens. Lower maximum thermal imaging readings were also recorded for the composite in all milling variants. The findings provide a qualitative assessment of machinability that takes account of differences in the forming methods and geometries of the compared specimens.

The results supported the central hypothesis within the scope of the experiments performed. A PLA/E90/CP composite with reduced combustion intensity and more favourable machining behaviour was developed, and relationships between raw material preparation, extrusion conditions and material properties were established. These findings provide a basis for further development of manufacturing technology for PLA-based components requiring machining after forming.