Machine Learning-guided Design of Organic Phosphorus-containing Flame Retardants to Improve the Limiting Oxygen Index of Epoxy Resins

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INTRODUCTION

- **Epoxy resin (EP)**, one of the most important polymer materials, has helped the rapid development of **microelectronics, energy, and space technology**[1]. However, the **inherent flammability** of EPs has severely limited their more extensive applications[2].
- The addition of **organic phosphorus-containing flame retardants (OPFRs)** by physical methods has **improved** the **fire resistance** of EPs, especially by increasing the **limiting oxygen index (LOI)**.
- The **development** of EP/OPFR composites is often through **trial and error**, with **no systematic understanding** on the OPFR **structure** and added **amount**.

OBJECTIVES

- Development of data-driven **machine learning (ML)** model for quantitative relationship between the **composite properties and the OPFRs structure and its content** in EP composites
- Using the ML model to **guide the design of new OPFRs and the amount to be added** for high performance EP composites

MACHINE LEARNING CONSTRUCTION

Figure 1. The database and ML construction steps.

- **Database construction:** 1053 data points were extracted manually from over 300 published papers.
- **ML Model development:** All ML programs were implemented through the Python package scikit-learn[3].

MATERIALS AND METHODS

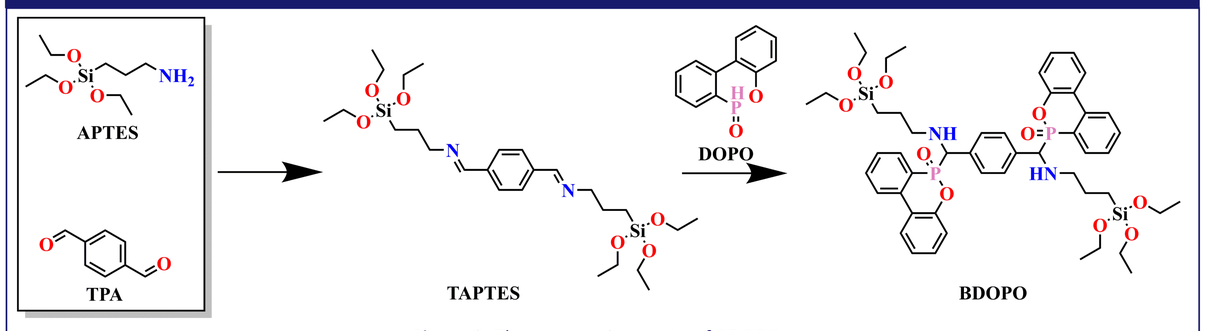

Figure 2. The preparation route of BDOPO.

- **A new FR – BDOPO, with the structure predicted from the ML model was synthesized experimentally.**
- **Structure:** Fourier transform infrared (FTIR) spectroscopy and ^{1}H magnetic resonance (^{1}H NMR) spectra.
- **Morphology:** Transmission electron microscopy (TEM) and scanning electron microscopy (SEM).
- **Flame retardancy:** LOI tests were measured on a JF-3 oxygen index tester.

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RESULTS AND DISCUSSIONS

1. Model performance

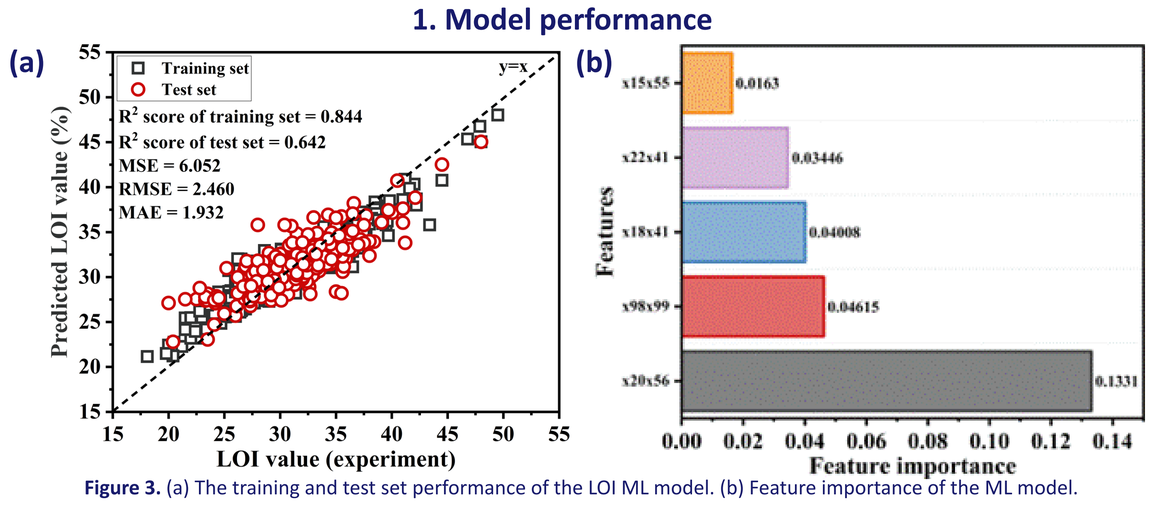

Figure 3. (a) The training and test set performance of the LOI ML model. (b) Feature importance of the ML model.

2. Structure of BDOPO

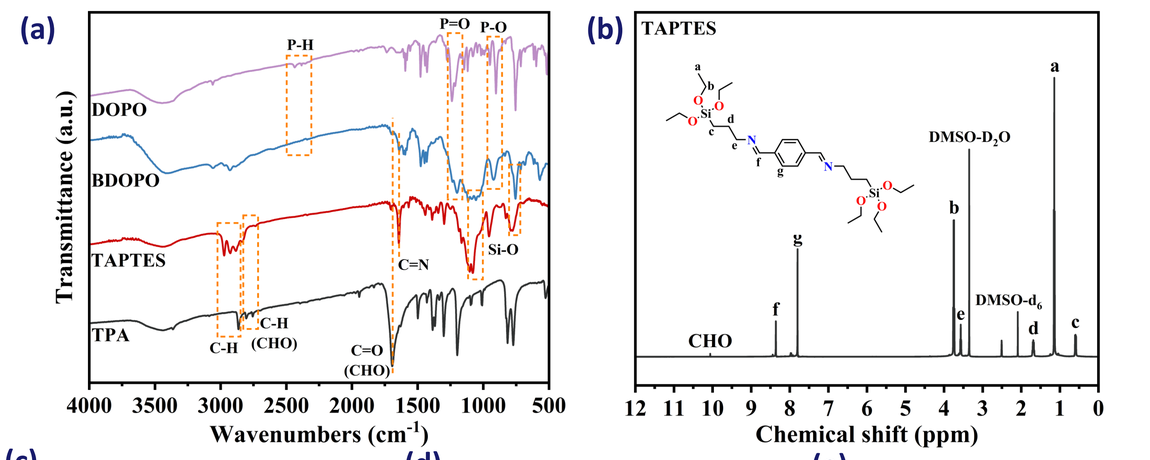

Figure 4. (a) FTIR of spectra. (b) ^{1}H NMR spectra. (c) SEM image of TAPTES. (d) and (e) SEM and TEM image of BDOPO.

3. Model Prediction and Validation

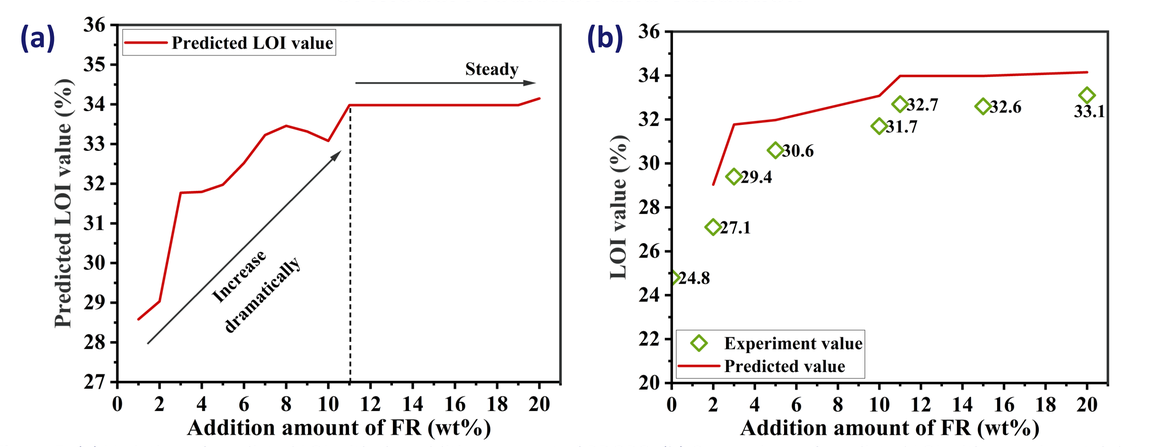

Figure 5. (a) Variation of predicted LOI with the addition amount of BDOPO. (b) Comparison of predicted LOI with experimental data.

KEY FINDINGS AND CONCLUSIONS

- The **coefficient of determination (R^2)** score of the training and test set is **0.844** and **0.642**, respectively (Fig. 3a).
- The trained ML model indicated that **conjugated systems (x20), phosphate esters (x56 and x98), nitrogen elements (x15 and x41) and addition amount (x99)** of OPFRs were also highly related to the LOI of EPs (Fig. 3b).
- The BDOPO was **synthesized** under the **guidance** of the ML model (Fig. 4).
- The **predicted LOI increases** with the **addition amount** of BDOPO up to 11% and then becomes steady (Fig. 5a).
- The **average error** between **prediction** and **experiments** was **5.1%** (Fig. 5b).
- The **ML** shows great potential in guiding the **design** of new OPFRs and **the amount to be added** for high performance **EP** composites.

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