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Advanced design of novel benzoxazines via a materials genome approach

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ABSTRACT

Achieving a precise balance between performance and the development of new benzoxazines poses a significant challenge. This article introduces a computer-aided screening method based on the materials genomes approach. This method enables the high-throughput design of molecular benzoxazine structures and the rapid screening of compounds that meet specific application requirements. It is subsequently applied to the development of new benzoxazines, which are anticipated to exhibit high-temperature resistance, low dielectric constants, and ease of processing. A benzoxazine with the best comprehensive performance is identified through screening 1122 designed structures, obtained by combining randomly selected 11 amines and 102 phenols. The experimental results show that the Td_5 of this benzoxazine is 410 °C, the dielectric constant is 2.9 (at 1 MHz), and the dielectric loss is 0.008. It also exhibits excellent processability, with a melting point of 78 °C, a processing temperature window of 80 °C, and a minimum viscosity of only 5.4 Pa s. It also surpasses most traditional benzoxazine in terms of heat resistance, dielectric properties, and processing performance, demonstrating the effectiveness of the materials genome method in developing new benzoxazines. Compared with traditional trial and error methods, the materials genome approach is more targeted and accurate, which enables the rapid and efficient development of benzoxazines.

1. Introduction

Polybenzoxazine is a relatively new member of the high-performance polymer materials family. In recent years, polybenzoxazine have received considerable attention due to their attractive properties, such as the absence of small molecule release during the curing process [1,2], low curing shrinkage [3], catalyst-free curing cross-linking [4–6], good mechanical properties [7], excellent dielectric properties [8,9], high heat resistance and residual carbon rate [10], low water absorption [11], and flexible molecular design [12–14]. As an outstanding thermosetting resin, benzoxazine is currently one of the research hotspots in thermosetting resins [15–19]. It is widely used in many fields, such as supercapacitor materials [20] and aerospace composites [21–23].

However, further improvement is still required to meet the multi-faceted requirements of aerospace applications [24], especially in terms of thermal stability and dielectric properties. In addition, considerable research has focused on the modification of benzoxazines. Functional groups such as cyano [25–28], fluorine-containing groups [29], and siloxanes [30,31] have been introduced into the structure of benzoxazine to enhance its thermal stability and/or dielectric properties. However, in most cases, the thermal stability of benzoxazines is improved at the expense of the processing properties. Balancing heat resistance, dielectric properties, and processing properties remains a significant challenge in the development of new benzoxazines.

Traditional research methods are mainly trial-and-error method, which is time-consuming and inefficient. The emergence of the materials genome approach offers the possibility of solving this problem. The materials genome approach adopts the research strategy of prioritising theoretical simulation guidance followed by experimental verification, aiming to accelerate the development of new materials. In recent years, there has been an increasing number of reports on the design of novel

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