

# Graphene-Like Materials from Biomass Using Deep Eutectic Solvents: A Review

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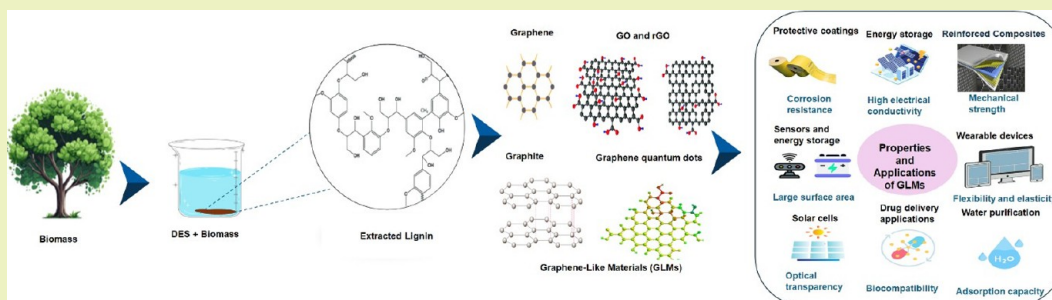
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**ABSTRACT:** Material sustainability is an ongoing challenge, and their renewable sourcing is the ultimate solution. Graphene-like materials (GLMs) such as graphene, graphene oxide, reduced graphene oxide, graphene quantum dots, and graphite are home to enormous physical and chemical properties exploitable for a range of applications. Lignin, a major component in plant biomass, shares structural similarity with GLMs and, therefore, could be their renewable source. The focus of this work is on the methods employed for the extraction of lignin from biomass using deep eutectic solvents (DESs). DESs have proven to be efficient in the isolation of lignin, presenting a sustainable pathway for the production of GLMs. Results from various studies are presented to demonstrate how lignin can be converted to GLMs. The implications of these findings extend beyond material sustainability and include applications in various fields, such as electronics and energy storage devices. This Review not only addresses the existing knowledge but also contributes to the advancement of ecofriendly methodologies in the pursuit of GLMs, thereby fostering material sustainability.

**KEYWORDS:** deep eutectic solvents, graphene-like materials, lignin, plant biomass, chemical processing

## 1. INTRODUCTION

Material sustainability has become the dominant topic in modern life due to increased greenhouse gas emissions associated with the sourcing and processing of conventional earthborn materials besides their overconsumption.<sup>1–3</sup> In this regard, lignocellulosic biomass (LCB) is a potentially sustainable and carbon-negative precursor for both energy and chemical production.<sup>4–6</sup> LCB is a blend of organic molecules consisting of oxygen, hydrogen, nitrogen, carbon, sulfur, and phosphorus as well as alkali and alkaline-earth metals.<sup>7</sup> LCB is Earth's most abundant, carbon-negative, renewable, and economical resource but intricately occurs with a complex network of three biopolymers: cellulose (30–40 wt %), hemicellulose (20–30 wt %), and lignin (10–25 wt %) (Figure 1). A compilation of these compositions for several biomasses is shown in Table S1. This complexity significantly hinders the efficient utilization of lignocellulosic feedstock.<sup>8,9</sup> Cellulose fibers aggregate into a bundle of microfibrils embedded in a matrix of hydrated polysaccharides (Figure 1d). Both cellulose and hemicellulose are polysaccharides; cellulose is formed by polymerization by 1,4-glycosidic linkages of  $\beta$ -glucose (Figure 1e,f), while hemicellulose is a hetero-

polymer composed of five and six carbon sugars, e.g., glucose, galactose, mannose, arabinose, and xylose (Figure 1g,h), and both are feedstocks for bioethanol, biobutanol, or fuels.<sup>10,11</sup> On the other hand, lignin exhibits a three-dimensional (3D) structure composed of phenylpropane building units, coniferyl alcohol, sinapyl alcohol, and paracoumaryl alcohol (Figure 1i,j) with carbon contents of >60%, high aromaticity, high oxygen functionalities, and tunable structure.<sup>12,13</sup>

Lignin offers enormous possibilities to manufacture graphene-like materials (GLMs) because of its structural similarity. Graphene is a multifunctional material having a one-atom-layer-thick sheet of  $sp^2$ -hybridized carbons in a honeycomb lattice with a domain-size-dependent electrical conductivity that can reach up to 80 MS/m<sup>14</sup> and mechanical

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