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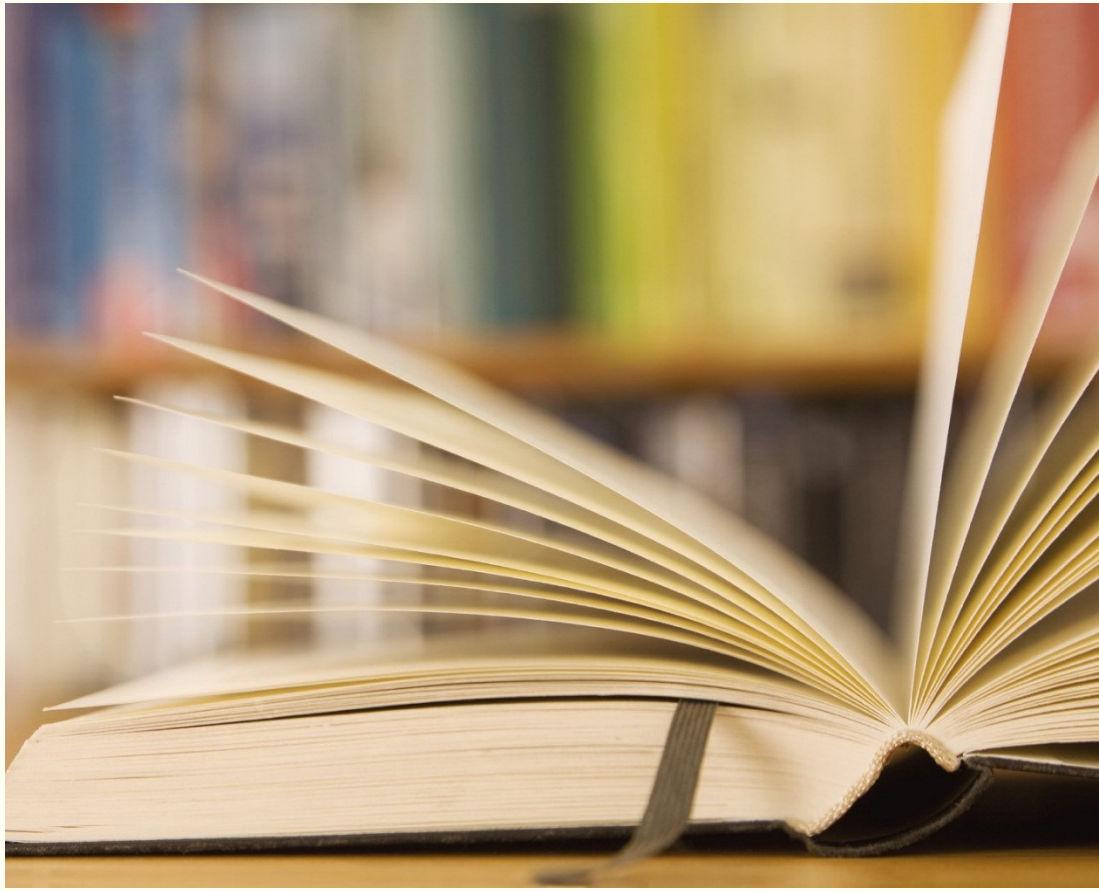

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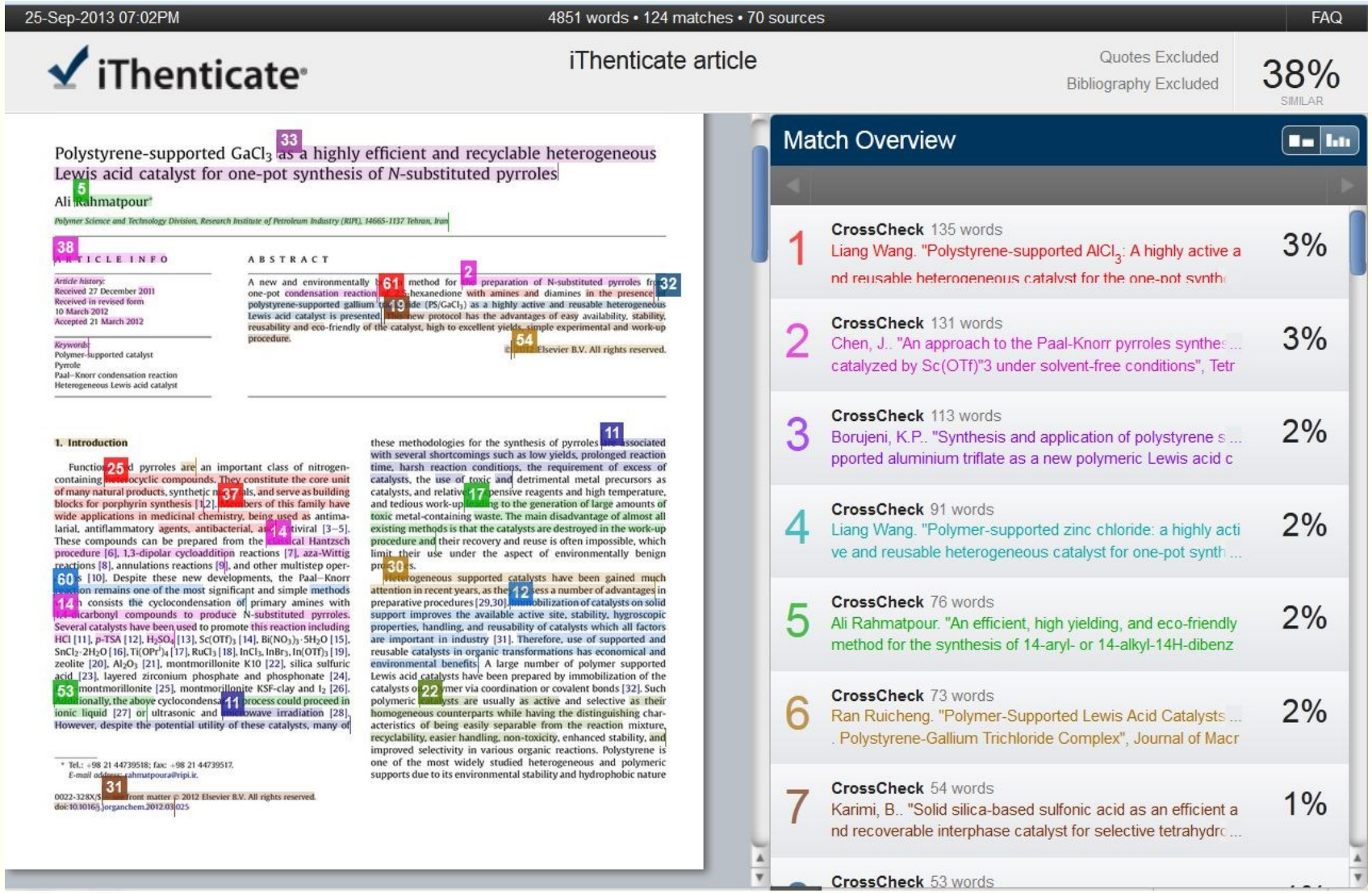
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Polystyrene-supported GaCl3 as a highly efficient and recyclable heterogeneous Lewis acid catalyst for one-pot synthesis of N-substituted pyrroles
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ABSTRACT
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A new and environmentally benign method for the preparation of N-substituted pyrroles from one-pot condensation reaction of 2,5-hexanedione with amines and diamines in the presence of polystyrene-supported gallium trichloride (PS/GaCl3) as a highly active and reusable heterogeneous Lewis acid catalyst is presented. The new protocol has the advantages of easy availability, stability, reusability and eco-friendly of the catalyst, high to excellent yields, simple experimental and work-up procedure.
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1. Introduction
Functionalized pyrroles are an important class of nitrogen-containing heterocyclic compounds. They constitute the core unit of many natural products, synthetic materials, and serve as building blocks for porphyrin synthesis [1,2]. Members of this family have wide applications in medicinal chemistry, being used as antimalarial, antiflammatory agents, antibacterial, and antiviral [3–5]. These compounds can be prepared from the classical Hantzsch procedure [6], 1,3-dipolar cycloaddition reactions [7], aza-Wittig reactions [8], annulations reactions [9], and other multistep operations [10]. Despite these new developments, the Paal–Knorr reaction remains one of the most significant and simple methods which consists the cyclocondensation of primary amines with 1,4-dicarbonyl compounds to produce N-substituted pyrroles. Several catalysts have been used to promote this reaction including HCl [11], p-TSA [12], H2SO4 [13], Sc(OTf)3 [14], Bi(NO3)3·5H2O [15], SnCl2·2H2O [16], Ti(OPr)4 [17], RuCl3 [18], InCl3, InBr3, In(OTf)3 [19], zeolite [20], Al2O3 [21], montmorillonite K10 [22], silica sulfuric acid [23], layered zirconium phosphate and phosphonate [24], montmorillonite [25], montmorillonite KSF-clay and I2 [26]. Additionally, the above cyclocondensation process could proceed in ionic liquid [27] or ultrasonic and microwave irradiation [28]. However, despite the potential utility of these catalysts, many of these methodologies for the synthesis of pyrroles are associated with several shortcomings such as low yields, prolonged reaction time, harsh reaction conditions, the requirement of excess of catalysts, the use of toxic and detrimental metal precursors as catalysts, and relatively expensive reagents and high temperature, and tedious work-up leading to the generation of large amounts of toxic metal-containing waste. The main disadvantage of almost all existing methods is that the catalysts are destroyed in the work-up procedure and their recovery and reuse is often impossible, which limit their use under the aspect of environmentally benign processes.
Heterogeneous supported catalysts have been gained much attention in recent years, as they possess a number of advantages in preparative procedures [29,30]. Immobilization of catalysts on solid support improves the available active site, stability, hygroscopic properties, handling, and reusability of catalysts which all factors are important in industry [31]. Therefore, use of supported and reusable catalysts in organic transformations has economical and environmental benefits. A large number of polymer supported Lewis acid catalysts have been prepared by immobilization of the catalysts on polymer via coordination or covalent bonds [32]. Such polymeric catalysts are usually as active and selective as their homogeneous counterparts while having the distinguishing characteristics of being easily separable from the reaction mixture, recyclability, easier handling, non-toxicity, enhanced stability, and improved selectivity in various organic reactions. Polystyrene is one of the most widely studied heterogeneous and polymeric supports due to its environmental stability and hydrophobic nature
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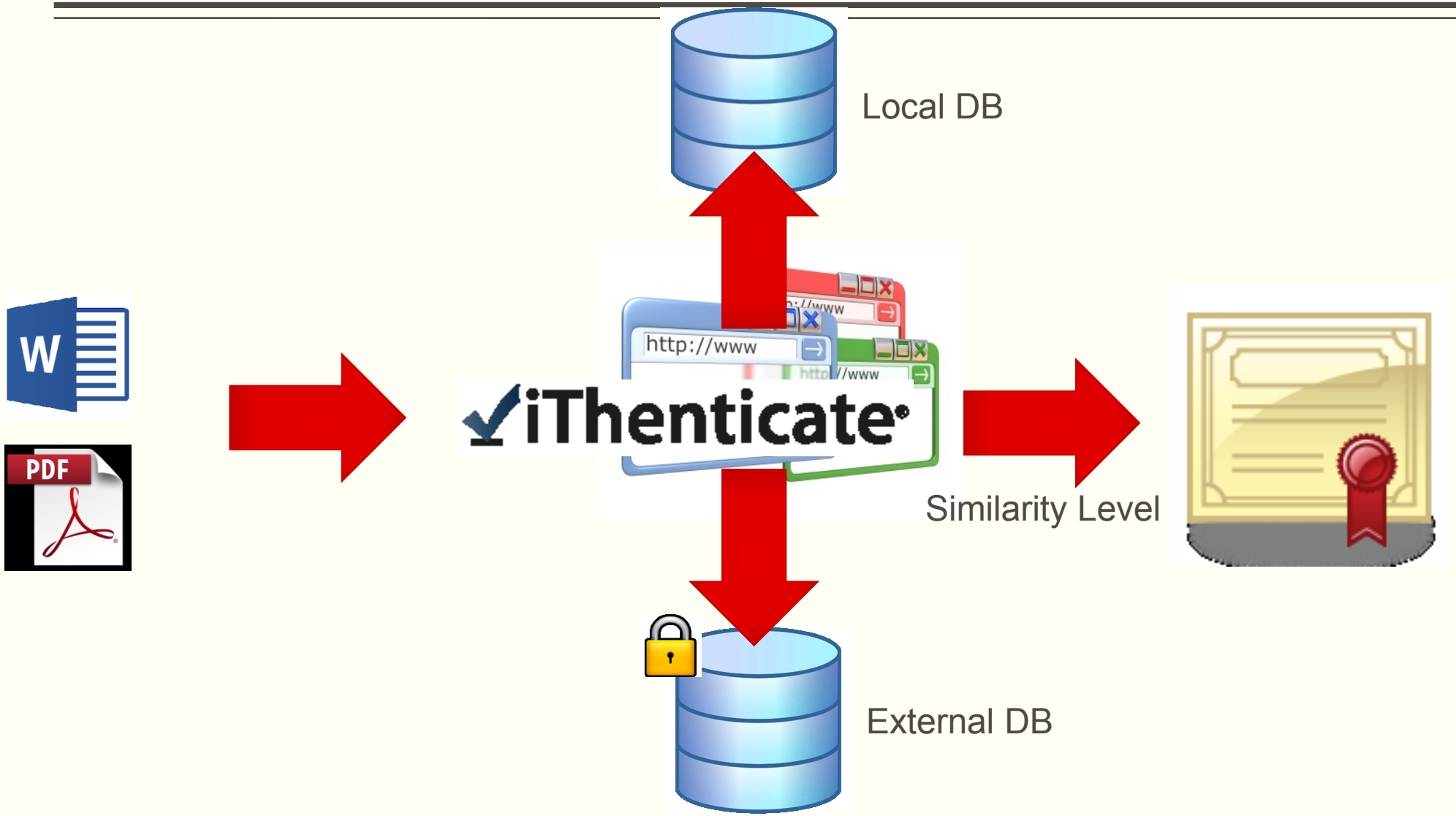

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