

PREPARASI ASAM FOSFOMOLIBDAT TERENKAPSULASI DALAM KERANGKA LOGAM-ORGANIK HKUST-1 SEBAGAI KATALIS HETEROGEN PADA SINTESIS TURUNAN FENILHIDRAZON

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INTISARI

Preparasi polioksometalat asam fosfomolibdat (PMA) terenkapsulasi dalam kerangka logam-organik (MOF) berbasis logam tembaga dengan ligan asam-1,3,5-benzena trikarboksilat yaitu Hong Kong University of Science and Technology-1 (HKUST-1) sebagai katalis reaksi sintesis turunan fenilhidrazon telah dilakukan. Penelitian ini bertujuan mendapatkan dan mengkarakterisasi katalis PMA@HKUST-1 serta mengevaluasi kinerjanya pada sintesis turunan fenilhidrazon.

Sintesis komposit PMA@HKUST-1 dilakukan secara *in situ* melalui metode *bottle around ship* dengan mereaksikan 1,0 mmol tembaga(II) nitrat trihidrat, 0,6 mmol asam-1,3,5-benzena trikarboksilat, dan beberapa variasi PMA meliputi: 0,04; 0,08; 0,16; dan 0,24 mmol dalam campuran pelarut metanol dan air (1:1 v/v) pada temperatur ruang selama 6 jam. Material hasil sintesis dikarakterisasi dengan XRD, FTIR, SAA, dan SEM-EDX. Selanjutnya, model reaksi antara 1,0 mmol benzaldehida dan 1,0 mmol fenilhidrazin digunakan mengoptimasi kondisi reaksi sintesis fenilhidrazon yang meliputi jenis pelarut, massa katalis, variasi katalis, dan metode sintesis. Kondisi optimum yang diperoleh diaplikasikan dalam sintesis turunan fenilhidrazon menggunakan variasi substrat benzaldehida, 4-bromobenzaldehida, isatin, dan 5-nitroisatin dengan jumlah yang sama yaitu 1,0 mmol. Produk turunan fenilhidrazon tersebut dikarakterisasi dengan uji titik lebur, serta spektrometer FTIR, dan GC-MS. Selain itu, stabilitas kinerja material sebagai katalis dievaluasi melalui uji penggunaan kembali sebanyak empat siklus reaksi yang dilanjutkan dengan karakterisasi pascareaksi menggunakan FTIR.

Hasil karakterisasi material katalis mengindikasikan bahwa PMA telah terenkapsulasi di dalam pori HKUST-1. Enkapsulasi PMA tidak menyebabkan perubahan struktur HKUST-1 yang dapat dilihat dari parameter kisi hasil Le Bail *refinement*. Sementara itu, kondisi reaksi optimum sintesis fenilhidrazon tercapai menggunakan 20 mg katalis PMA(0,16)@HKUST-1 dalam pelarut etanol dengan metode sonikasi pada suhu 50 °C selama 2 jam. Katalis tersebut dapat mengkatalisis reaksi kondensasi dan menghasilkan turunan fenilhidrazon dengan perolehan persen hasil berkisar antara 87% hingga 93%. Selain itu, hasil uji penggunaan kembali katalis menunjukkan bahwa hingga empat siklus reaksi kinerjanya tetap terjaga dengan persen hasil mencapai 91%.

Kata kunci: enkapsulasi, fenilhidrazon, HKUST-1, katalis, PMA

PREPARATION OF PHOSPHOMOLYBDIC ACID ENCAPSULATED IN HKUST-1 METAL-ORGANIC FRAMEWORK AS HETEROGENEOUS CATALYSTS FOR SYNTHESIS PHENYLHYDRAZONE DERIVATIVES

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ABSTRACT

Preparation of a polyoxometalate, phosphomolybdic acid (PMA) encapsulated within a copper-based metal-organic framework (MOF) with 1,3,5-benzene tricarboxylic acid ligand, Hong Kong University of Science and Technology-1 (HKUST-1) as a catalyst for the synthesis of phenylhydrazone derivatives was carried out. The objective of this research was to prepare and characterize the PMA@HKUST-1 catalyst, as well as to evaluate its catalytic performance in the synthesis of phenylhydrazone derivatives.

The synthesis of the PMA@HKUST-1 composite was performed *in situ* via the bottle around ship method by reacting 1.0 mmol copper(II) nitrate trihydrate, 0.6 mmol 1,3,5-benzene tricarboxylic acid, and various amounts of PMA (0.04, 0.08, 0.16, and 0.24 mmol) in a mixture of methanol and water solvents (1:1 v/v) at room temperature for 6 hours. The synthesized materials were characterized using XRD, FTIR, SAA, and SEM-EDX. Furthermore, a model reaction between 1.0 mmol benzaldehyde and 1.0 mmol phenylhydrazine was utilized to optimize the reaction conditions for phenylhydrazone synthesis, including solvent type, catalyst mass, catalyst variation, and synthesis method. The optimum conditions obtained were applied to the synthesis of phenylhydrazone derivatives using substrate variations of benzaldehyde, 4-bromobenzaldehyde, isatin, and 5-nitroisatin at the same amount of 1.0 mmol. The phenylhydrazone derivatives were characterized by melting point determination, FTIR, and GC-MS spectrometry. In addition, the catalytic stability of the material was evaluated through reusability tests over four reaction cycles, followed by post-reaction characterization using FTIR.

The characterization results of the catalyst material indicated that PMA was encapsulated within the pores of HKUST-1. The encapsulation of PMA did not induce structural changes in HKUST-1, as evidenced by the lattice parameters obtained from Le Bail refinement. Meanwhile, the optimum reaction conditions for phenylhydrazone synthesis were achieved using 20 mg of PMA(0.16)@HKUST-1 catalyst in ethanol solvent under ultrasound irradiation at 50 °C for 2 hours. The catalyst was capable of catalyzing the condensation reaction for the synthesis phenylhydrazone derivatives with yields ranging from 87% to 93%. Moreover, the reusability test results demonstrated that its performance remained stable for up to four reaction cycles, with a yield reaching 91%.

Keywords: catalyst, encapsulation, HKUST-1, phenylhydrazone, PMA