INTISARI

Permasalahan peningkatan emisi karbon dioksida (CO_2) akibat aktivitas industri dan penggunaan energi fosil menjadi isu global yang mendesak. *Graphene oxide* (GO) sebagai salah satu material berbasis karbon memiliki potensi besar dalam teknologi adsorpsi CO_2 karena kaya akan gugus fungsional oksigen. Pemanfaatan grafit dari limbah baterai NMC (*Lithium Nickel Cobalt Manganese Oxide*) sebagai bahan baku *graphene oxide* (GO) menawarkan pendekatan yang lebih berkelanjutan sekaligus bernilai ekonomis, namun karakteristik kimia dan struktur GO yang dihasilkan sangat dipengaruhi oleh kondisi pra-oksidasi dan oksidasi selama sintesis. Permasalahan utama dalam penelitian ini adalah bagaimana meningkatkan kandungan gugus fungsional khususnya gugus karboksil, yang penting untuk aplikasi adsorpsi CO_2 maupun aplikasi fungsional lainnya, serta bagaimana mengoptimalkan kondisi operasi agar menghasilkan GO dengan tingkat oksidasi dan struktur lapisan yang stabil.

Penelitian ini bertujuan untuk menganalisis karakteristik GO yang disintesis menggunakan grafit limbah baterai NMC melalui modifikasi metode Hummer, mengoptimasi pra-oksidasi menggunakan hidrogen peroksida untuk meningkatkan rasio gugus karboksil, serta menentukan pengaruh variasi suhu dan waktu oksidasi terhadap kualitas dan struktur GO yang dihasilkan. Proses sintesis dimulai dengan tahap pra-oksidasi menggunakan H_2O_2 pada tiga variasi rasio (15, 25 dan 35 mL/g grafit), kemudian dilanjutkan dengan sintesis GO menggunakan modifikasi metode Hummer dengan variasi suhu (30, 45 dan 60°C) dan waktu oksidasi (2, 5 dan 8 jam) sesuai rancangan percobaan. Karakterisasi meliputi analisis Raman Spectroscopy, FTIR, XRD, TGA, N_2 Sorption SEM, titrasi Boehm, XPS dan kapasitas penjerapan CO_2. Data dianalisis menggunakan *Response Surface Methodology* (RSM) untuk menemukan kombinasi kondisi operasi yang menghasilkan konsentrasi gugus karboksil optimal.

Hasil penelitian menunjukkan bahwa pra-oksidasi menggunakan H_2O_2 berperan penting dalam membuka struktur grafit dan memfasilitasi pembentukan gugus oksigen. Namun rasio H_2O_2 yang terlalu tinggi (35 mL/g) cenderung menyebabkan degradasi struktur awal grafit sehingga menurunkan efektivitas oksidasi pada tahap berikutnya. Variasi suhu dan waktu oksidasi menunjukkan pola peningkatan gugus karboksil hingga kondisi optimum, kemudian menurun akibat terjadinya dekarboksilasi dan fragmentasi lembaran pada kondisi oksidatif yang terlalu kuat. Analisis kontur dan permukaan respon menunjukkan adanya interaksi signifikan antara rasio H_2O_2, suhu dan waktu oksidasi terhadap karakteristik GO.

Kondisi optimum diperoleh pada pra-oksidasi dengan 25 mL/g H_2O_2, suhu oksidasi 45°C dan waktu oksidasi 5 jam, yang menghasilkan GO dengan konsentrasi gugus karboksil 6,5 mmol/g, karakteristik struktural yang konsisten dengan tingkat oksidasi yang lebih tinggi, stabilitas termal khas GO teroksidasi, serta kapasitas adsorpsi CO_2 sebesar 0,02 mmol/g. Secara keseluruhan, penelitian ini berhasil menunjukkan bahwa kombinasi pra-oksidasi H_2O_2 dan optimasi kondisi operasi mampu meningkatkan sifat kimia dan struktur GO berbasis limbah baterai NMC.

Kata kunci: *graphene oxide*; baterai NMC; pra-oksidasi hidrogen peroksida; *Hummers method*; gugus karboksil.

ABSTRACT

The increasing emission of carbon dioxide (CO_2) resulting from industrial activities and fossil energy consumption had become a pressing global issue. Graphene oxide (GO), as a carbon based material, had shown great potential for CO_2 adsorption technologies due to its high density of oxygen-containing functional groups. The utilization of graphite derived from spent lithium nickel cobalt manganese oxide (NMC) batteries as a precursor for graphene oxide offered a more sustainable and economically valuable approach. However, the chemical characteristics and structural properties of the resulting GO were strongly influenced by the pre-oxidation and oxidation conditions during synthesis. The main challenges addressed in this study were how to enhance the content of functional groups, particularly carboxyl groups, which are crucial for CO_2 adsorption and other functional applications, and how to optimize the operating conditions to produce GO with a stable oxidation level and layered structure.

This study aimed to analyze the characteristics of GO synthesized from NMC battery waste graphite through a modified Hummers method, to optimize hydrogen peroxide pre-oxidation in order to increase the carboxyl group ratio, and to determine the effects of oxidation temperature and time on the quality and structure of the resulting GO. The synthesis process began with a pre-oxidation step using H_2O_2 at three different ratios (15, 25, and 35 mL/g of graphite), followed by GO synthesis via a modified Hummers method with variations in oxidation temperature (30, 45, and 60 °C) and oxidation time (2, 5, and 8 h) according to the experimental design. The synthesized GO was characterized using Raman spectroscopy, Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), thermogravimetric analysis (TGA), nitrogen adsorption–desorption (N_2 sorption), scanning electron microscopy (SEM), Boehm titration, X-ray photoelectron spectroscopy (XPS), and CO_2 adsorption capacity measurements. The experimental data were analyzed using Response Surface Methodology (RSM) to identify the optimal combination of operating conditions that produced the highest carboxyl group concentration.

The results indicated that pre-oxidation using H_2O_2 played a crucial role in opening the graphite structure and facilitating the formation of oxygen containing functional groups. However, an excessively high H_2O_2 ratio (35 mL/g) tended to cause degradation of the initial graphite structure, thereby reducing the effectiveness of subsequent oxidation. Variations in oxidation temperature and time exhibited an increasing trend in carboxyl group content up to an optimum condition, followed by a decrease due to decarboxylation and sheet fragmentation under overly strong oxidative conditions. Contour and response surface analyses revealed significant interactions among the H_2O_2 ratio, oxidation temperature, and oxidation time in determining GO characteristics.

The optimum condition was obtained at a pre-oxidation ratio of 25 mL/g H_2O_2, an oxidation temperature of 45 °C, and an oxidation time of 5 h. Under these conditions, the synthesized GO exhibited a carboxyl group concentration of 6.5 mmol/g, structural characteristics consistent with a higher degree of oxidation, thermal stability typical of oxidized GO, and a CO_2 adsorption capacity of 0.02 mmol/g. Overall, this study successfully demonstrated that the combination of H_2O_2 pre-oxidation and optimized operating conditions enhanced the chemical properties and structural characteristics of GO derived from NMC battery waste.

Keywords: graphene oxide; NMC battery; hydrogen peroxide pre-oxidation; Hummer method; carboxyl group.