

**Preparation and Characterization of Disodium Terephthalate/Graphene Composites**  
**การเตรียมและพิสูจน์เอกลักษณ์ของวัสดุคอมโพสิตระหว่างไดโซเดียมเทเรฟทาเลตและกราฟีน**

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**ABSTRACT**

Currently, a demand on lithium-ion batteries (LIBs) to store electric energy increases continuously. Even though they are widely used today, the lithium ores decrease and leads to limit the production of LIBs. Sodium, an IA element is a good candidate to be used instead of lithium because it is abundant, inexpensive and non-toxic. In this work, disodium terephthalate and graphene were prepared. Disodium terephthalate was derived from terephthalic acid and sodium hydroxide. Graphene was obtained from the hydrothermal process of table sugar. SEM images and XRD pattern confirm the successful preparation of disodium terephthalate and graphene. The sharp crystals of disodium terephthalate were ~50 microns and the spherical powder of graphene was also ~50 microns.

**บทคัดย่อ**

ปัจจุบันนี้ความต้องการลิเทียมไอออนแบตเตอรี่ในการจัดเก็บพลังงานไฟฟ้ามีเพิ่มมากขึ้นอย่างต่อเนื่อง ถึงแม้ว่าแบตเตอรี่เหล่านี้จะมีการใช้กันอย่างแพร่หลาย แวลลิเทียมจึงมีปริมาณที่ลดลงและนำไปสู่การจำกัดการผลิตของลิเทียมไอออนแบตเตอรี่ โซเดียมเป็นธาตุหมู่หนึ่งเอซึ่งมีคุณสมบัติที่ดีพอที่จะนำมาใช้แทนลิเทียมได้เพราะในธรรมชาติมีโซเดียมอยู่มาก ราคาไม่แพงและไม่เป็นพิษ ในงานวิจัยนี้ไดโซเดียมเทเรฟทาเลตและกราฟีนได้เตรียมขึ้นโดยไดโซเดียมเทเรฟทาเลตได้จากกรดเทเรฟทาลิกและโซเดียมไฮดรอกไซด์ ส่วนกราฟีนเตรียมจากกระบวนการไฮโดรเทอร์มอลของน้ำตาลทราย ภาพ SEM และ รูปแบบ XRD ยืนยันการประสบความสำเร็จในการเตรียมไดโซเดียมเทเรฟทาเลตและกราฟีน ผลึกที่แหลมคมของไดโซเดียมเทเรฟทาเลตมีขนาดประมาณ 50 ไมครอนและผงกลมๆของกราฟีนก็มีขนาดประมาณ 50 ไมครอนเช่นกัน

**Key Words:** Composite, Disodium terephthalate, Graphene

**คำสำคัญ:** คอมโพสิตไดโซเดียมเทเรฟทาเลตกราฟีน

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## Introduction

Fossil fuels, which are coal, petroleum and natural gas, have been important energy sources in the world for several hundred years ago. Unfortunately, the demand on energy still increases. In addition, fossil fuels are non-renewable energy and cause environmental hazards. Presently, there are several attempts to generate the renewable and sustainable energy from various sources such as sunlight, wind, water and biomass. To store the produced electricity, rechargeable batteries are required (Reinhard et al., 2013 and Zhenguot et al., 2011).

Among them, rechargeable lithium ion batteries are the most widely used energy storage and were a rechargeable battery. Currently, the demand on lithium ion batteries increases, resulting in the reduction of lithium ores. This leads to the limitation on the production of lithium ion batteries.

Non-toxic Na-ion batteries are good candidates to be used as an alternative batteries (Zelang et al., 2013 and Hongli et al., 2014) because sodium is abundant, inexpensive and is produced from renewable resources.

There are several studies on the use of small organic molecules for storing either lithium or sodium ions. Although the production of these molecules use less energy than that of inorganic ones. They have several drawbacks such as poor recyclability and low thermal stability (Xin-Yun et al., 2007 and Thierry et al., 2003). Therefore, Tarascon and co-workers studied redox-active for carbonyl group-containing organic electrode materials in high performance lithium ion batteries (Makowski et al., 2009).

Terephthalic acid is also a good candidate to be used for storing sodium ions (Panasyuk et al., 2003). This chemical is normally obtained from recycling poly (ethylene terephthalate) (PET) waste bottles.

Graphene is an attractive candidate for improving electrical conductivity of these small organic molecules. Because of, graphene has surpassing electrical (Phaedon et al., 2007), thermal (Alexander et al., 2008) and mechanical properties (Dan et al., 2008).

Various methods have been used to prepare graphene such as mechanical exfoliation (Konstantin et al., 2004), graphite oxide reduction (Sungjin et al., 2009) and chemical vapor deposition (CVD) (Xuesong et al., 2009 and Libin et al., 2012).

In this work, graphene was synthesized from the hydrothermal process of table sugar and an abundant natural material. While Disodium terephthalate was obtained from the reaction between terephthalic acid and sodium hydroxide, a convenient process.

## Methodology

### Materials

The reagent grade commercial terephthalic acid powder (TPA 98%, Sigma-Aldrich Co. product of America), sodium hydroxide (NaOH 99%, AR Grade, RCI Labscan Co. product of Thailand) and ethanol (EtOH, AR Grade, RCI Labscan Co. product of Thailand) were used without further purification. The table sugar from local supply Thailand and deionized (DI) water were used to synthesis graphene.

### Synthesis and characterization of Na<sub>2</sub>TP and graphene

In typical preparation, 0.30 M of terephthalic acid and 0.63 M of sodium hydroxide were dissolved in ethanol (60 cc) together at room temperature for 24 hours. After that, the reaction mixture was centrifuged and the supernatant solution was decanted.

The sediment was redispersed in ethanol and centrifuged again for three times. The obtained white powder of Na<sub>2</sub>TP was dried in vacuum oven at 150 °C for 2

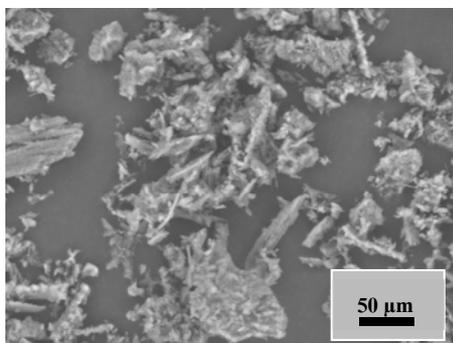
hours.

To obtain graphene, table sugar (0.8 M) was dissolved in deionized water (30 cc). The solution was poured into a Teflon-lined autoclave with incubation in vacuum oven at 220 °C for 24 hours.

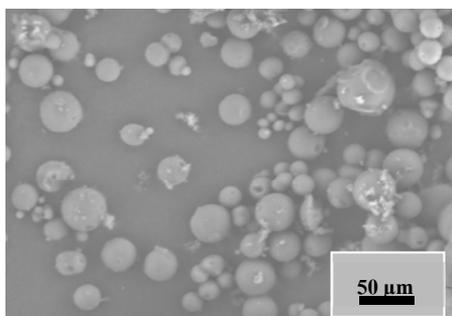
Both Na<sub>2</sub>TP and graphene were characterized by scanning electron microscopy (SEM, TM-1000 table Microscope product of Hitachi, Germany), x-ray diffraction (XRD, D8 Advance product of Bruker, United Kingdom), Fourier transform-infrared spectroscopy (FTIR, Nicolet 6700 product of Thermo scientific, United states) and thermogravimetric analysis (TGA, SDT 2960 Simultaneous DSC-TGA product of TA Instruments, United states).

### Results and discussion

The SEM images of Na<sub>2</sub>TP and graphene are shown in Figures 1 and 2, respectively.

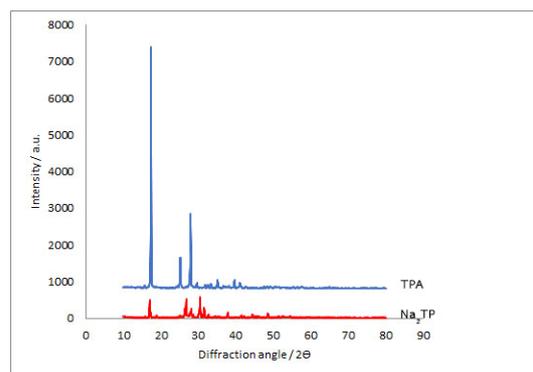


**Figure 1** A SEM image of Na<sub>2</sub>TP

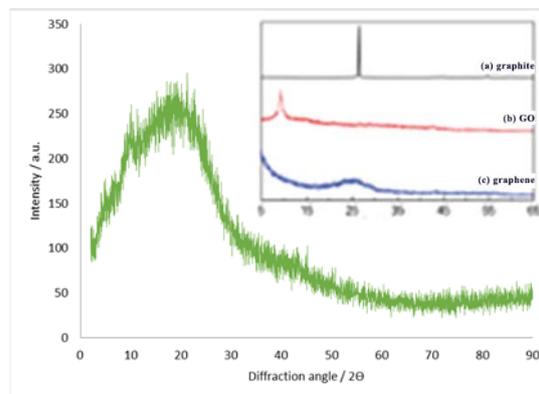


**Figure 2** A SEM image of graphene

The SEM image in Figure 1 presents the sharp surface of Na<sub>2</sub>TP crystals with the sizes of microns. Figure 2 shows the spherical graphene particles with the particle size of ~50 microns were spherical. The XRD pattern of Na<sub>2</sub>TP and graphene are shown in Figure 3 and Figure 4 respectively.



**Figure 3** XRD pattern of Na<sub>2</sub>TP



**Figure 4** XRD pattern of graphene.

The XRD pattern of Na<sub>2</sub>TP and graphene shown in Figure 3 and Figure 4 respectively. The XRD pattern of the products from the reaction between terephthalic acid and sodium hydroxide confirms the success of synthesis, when compared to XRD pattern of Na<sub>2</sub>TP (Yuwon et al., 2012). The XRD pattern of Na<sub>2</sub>TP had the shifting of peak for 2 $\theta$  is a higher than when compared to that of terephthalic acid. The XRD pattern of synthesized graphene from hydrothermal sugar shows a broad peak at ~10-25° while

that of graphene oxide does a sharp peak at 10°. The XRD pattern of graphite shows only one sharp peak at 25°. Therefore, it could be concluded that the product from the hydrothermal process of table sugar was graphene.

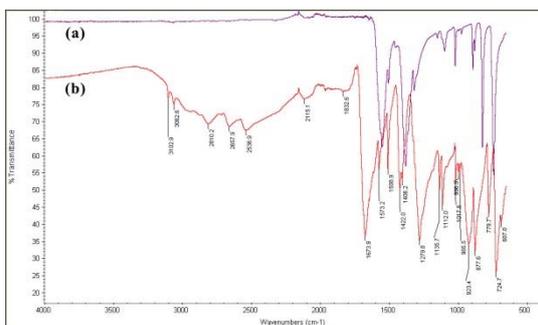


Figure 5 FTIR spectrum of Na<sub>2</sub>TP(a) and terephthalic acid (b)

The FTIR spectrum of Na<sub>2</sub>TP (a) in Figure 5 indicates metal carboxylate formation in product from the reaction between sodium hydroxide and terephthalic acid. The bands of Na<sub>2</sub>TP (a) at 1551 cm<sup>-1</sup> and 1380 cm<sup>-1</sup> were assigned to V<sub>s</sub>(COONa).Na<sub>2</sub>TP gave right hand-shift peaks compared to carboxylate peaks of terephthalic acid (b). The peaks of terephthalic acid (b) were centered at 3102 (OH stretching) and at 1673 and 1422 cm<sup>-1</sup> (C=O stretching). Also, the peak of OH stretching of carboxylic acid groups disappeared, indicating that the pure Na<sub>2</sub>TP was formed.

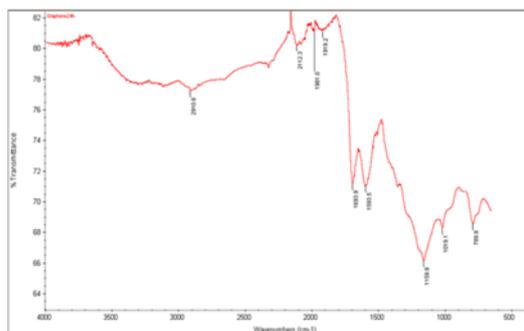


Figure 6 FTIR spectrum of graphene

Figure 6 shows the characteristic peaks of aromatic C=C vibration in the range of 1593-1693 cm<sup>-1</sup>. Upon reduction of graphene oxide to graphene the C=O stretching disappears and new band at 2910 cm<sup>-1</sup> was replaced the C-H stretch vibrations of the methylene group. Then, the FTIR spectrum confirms the graphene synthesis was successful.

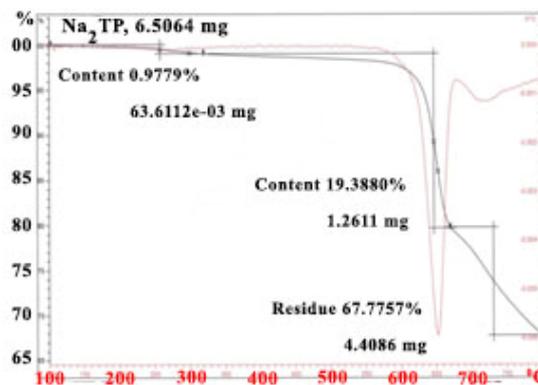


Figure 7 TGA curve of Na<sub>2</sub>TP

The TGA curve above shows two nodes at around 260 °C and 650 °C. TGA curve indicated that the product was a mixture of monosodium terephthalate (NaTP) and Na<sub>2</sub>TP, corresponding to the decomposition at 260 and 650 °C, respectively. However, when the obtained compounds were purified by recrystallization in ethanol, the thermogram shows only one sample loss near 650 °C, indicating the formation of pure Na<sub>2</sub>TP.

### Conclusion

Graphene and Na<sub>2</sub>TP were successfully synthesized from the hydrothermal process of table sugar and the reaction between terephthalic acid and sodium hydroxide.

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