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# Effects of Nickel Particle Size and Graphene Support on the Electrochemical Performance of Lithium/Dissolved Polysulfide Batteries



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#### ABSTRACT

The electrocatalytic effect of nickel (Ni) nanoparticle sizes on the lithium polysulfide conversion reactions in dissolved lithium sulfur battery configuration is investigated. The Ni particles of 20 nm with the higher cathode surface area show a superior capacity of  $1066 \, \text{mAh g}^{-1}$  sulfur compared to Ni particles of 40 and 100 nm for the first cycle. In addition, to further improve the capacity retention and discharge capacity of the cell, the effect of the graphene support on Ni nanoparticle dispersion and cycling performance is investigated. The results show a significant improvement in the discharge capacity compared to the other electrodes. This could be explained by the homogeneous distribution of Ni nanoparticle within the carbon matrix, which suppress the agglomeration and surface area loss of the Ni nanoparticle after cycling; as well as a synergetic effect of graphene structure and Ni nanoparticle.

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#### 1. Introduction

Advances in portable electronic devices and electric vehicles requires advanced electrical energy storage systems, such as rechargeable batteries with high energy density and long lasting cycle performance. Among the most promising energy storage systems, lithium sulfur (Li/S) batteries have received increasing attention due to their high energy density of 2600 Wh kg<sup>-1</sup> and theoretical capacity of  $1675 \,\mathrm{mAh}$  g<sup>-1</sup> [1]. In addition, sulfur is environmentally friendly and has a low cost [1-3]. However, the development of lithium sulfur batteries presents some obstacles such as poor cyclability, lower practical capacity and high selfdischarge rate [1-3]. The poor conductivity of sulfur and its final discharge products, Li<sub>2</sub>S<sub>2</sub> and Li<sub>2</sub>S, limits the specific capacity of Li/ S batteries. Furthermore, the dissolution of lithium polysulfide intermediates in the liquid electrolyte during the charge-discharge process causes the polysulfide shuttle process and lithium corrosion, which results in low cyclability, charging inefficiency and high self-discharging rate of batteries [4,5].

In recent years, efforts have been made to improve the performance of Li/S batteries and overcome the above challenges.

Much of the research has focused on the development of new cathode materials to enhance the electrical conductivity, to accommodate and absorb polysulfide and active materials within the pores or layers of the cathode structure. In order to improve cathode material performance, conductive carbon additives such as: mesoporous and nano-structured carbon [6–8], porous hollow carbon spheres [9,10], carbon multi-walled nano-tubes [11,12], graphene [13,14], conductive polymers [15–17], and carbon interlayers have been used. Several studies have been conducted on the design of alternative current collectors such as Ni foam as an interlayer, cathode conductive structure [18], and a protective layer [19], which improves the active material utilization, cycle life, and battery performance due to its metallicity and 3-D network structure.

It has been reported that the reaction kinetics of aqueous polysulfide can be enhanced by using electrocatalytic electrodes in photoelectrochemical solar cells [20] and redox flow battery cells [21]. Metal oxides such as  $CeO_2$  [22,23],  $MnO_2$  [24–26], have been demonstrated to be promising cathode catalysts for rechargeable batteries electrochemical reactions. In addition, some metal oxides such as  $TiO_2$  [27],  $Al_2O_3$  [28],  $SiO_2$  [29] and  $MnO_2$  [30] have been shown to enhance Li/S discharge capacities and cycling stability. This can be attributed to their strong ability to adsorb sulfur species, thus effectively suppressing the diffusion of polysulfide species into the electrolyte.  $Ti_4O_7$  has also been reported to

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enhance the Li/S batteries redox chemistry due to its sulphiphilic surface and good electron conductivity [5]. In our recent study, carbon-free electrocatalysts such as Pt, Au and Ni has been found to enhance lithium polysulfide conversion reactions and improve Li/S battery performance [31]. Among these electrocatalysts, Ni showed a promising behavior due to its high conductivity along with electrocatalytic activity toward polysulfide reduction reaction [31].

Very few studies have been conducted on lithium polysulfide dissolved configuration with electrocatalytic electrodes. Babu et al. [31] found that with a 200 nm thick Ni electrode and 10  $\mu$ l of 0.2 M Li<sub>2</sub>S<sub>8</sub> catholyte, a capacity of 420 mAh g<sup>-1</sup> sulfur was obobserved after 40 cycles; while Zhang et al. [32] measured a capacity of 490 mAh g<sup>-1</sup> sulfur with a super-p carbon electrode and 20  $\mu$ l of 0.25 m Li<sub>2</sub>S<sub>9</sub> catholyte, also after 40 cycles. In the present study, a series of Ni nanoparticles with nominal particle size of 20, 40, and 100 nm have been used as electrocatalysts, and the effect of particle size on Li/S battery performance has been investigated.

Furthermore, graphene has been chosen as a support to anchor the Ni nanoparticles and to investigate the synergetic effect of carbon material and Ni nanoparticles for the lithium polysulfide conversion reactions. The results indicated there is a strong particle size effect, and Ni/graphene electrode exhibits a capacity of 753 mAh  $\rm g^{-1}$  sulfur after 40 cycles which is significantly higher than similar studies [31,32].

#### 2. Experimental

#### 2.1. Preparation of electrocatalysts

Nickel nanoparticles (Ni, 99.9% with sizes of 20 nm, 40 nm and 100 nm, US Research Nanomaterial Inc.) were activated under hydrogen for 3 h at  $150\,^{\circ}$ C. To prepare Ni/graphene hybrids, 5 grams of graphene (Angstron Materials) were refluxed with 250 mL of 70% HNO<sub>3</sub> at  $110\,^{\circ}$ C for 3 h to functionalize and also remove metallic impurities. The material was then washed with water and

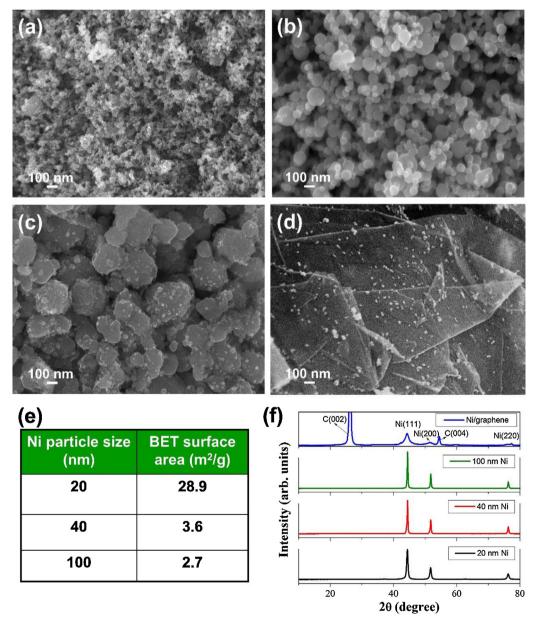


Fig. 1. SEM images of Ni nanoparticles: (a) 20 nm, (b) 40 nm, (c) 100 nm, (d) Ni/Graphene, (e) BET surface area of Ni nanoparticles, and (f) XRD patterns of all of the nanoparticles.

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