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Mixture of Carbon Derived from Lokta Paper and Graphite for Efficient Counter Electrodes of Dye-sensitized Solar Cells

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Abstract

This study explores the potential of using a mixture of Lokta paper-based activated carbon (80%) and graphite powder (20%) as an efficient counter electrode (CE) material for dye-sensitized solar cells (DSSCs). The activated carbon was synthesized in two steps. Firstly, Lokta paper was carbonized at 400 ºC. Then the char was activated with $ZnCl_2$ and carbonized at 800 ºC. The activated carbon and its mixture were characterized by the techniques of X-ray diffraction (XRD) and scanning electron microscope (SEM). The XRD of the activated carbon disclosed the formation of both graphitic and amorphous forms of carbon in the sample. The SEM image of the Lokta paper-derived activated carbon explored a rough and porous surface, and the Energy dispersive X-ray spectroscopy (EDS) of the mixture detected high carbon content in the sample. The catalytic ability of the mixture of the activated carbon and graphite for the reduction of tri-iodide ions was investigated by electrochemical impedance spectroscopy (EIS) of symmetrical dummy cells. The charge transfer resistance at the mixture-electrolyte interface of the symmetrical cell was 1.59 Ω-cm^2. Hence, the mixture of the Lokta paper-based activated carbon and graphite can be an efficient and low-cost CE material alternative to expensive platinum used in DSSCs.

Keywords: Activated carbon, counter electrode, dye-sensitized solar cells, graphite, Lokta paper, platinum

Introduction

Solar cells are devices which are designed to capture the solar photons and instantly convert them into electricity. Dye-sensitized solar cells (DSSCs) are comparatively a newer type of solar cell than crystalline silicon solar cells (Grätzel, 2003; Joshi et al., 2009; Poudel et al., 2012). A DSSC is a photochemical cell comprising photoelectrode, counter electrode (CE), and liquid electrolyte. The electrolyte contains iodide/tri-iodide ions, and it is sandwiched between the two electrodes.

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A typical photoelectrode is a fluorine-doped tin oxide (FTO) glass substrate coated with a dye-sensitized thin film of titanium dioxide (TiO_2). Similarly, CE is a platinized FTO-glass substrate. As light is incident on the photoelectrode, the dye molecules harvest light photons and inject their electrons into the conduction band of the TiO_2. The photoinduced electrons pass through a load in an external circuit and produce electric power, then the electrons arrive at the CE. On the other hand, the oxidized dye molecules are rejuvenated by receiving electrons from the iodide ions in the electrolyte. The iodide ions transform into tri-iodide ions after losing electrons. The tri-iodide ions gain the photoinduced electrons at the CE (Chaudhary et al., 2024; Grätzel, 2003, 2005; Joshi et al., 2020). A catalyst is coated at the CE to improve the rate of the charge transfer at the interface of the CE and electrolyte. Platinum (Pt) is widely used catalyst at the CEs of DSSCs, however, the high price of Pt is an issue (Joshi et al., 2020, 2025).

In the quest for a cheaper catalyst than Pt, researchers have proposed carbonaceous materials and their composites like carbon black (Murakami et al., 2006; Ramasamy et al., 2007), carbon nanotubes (Lee et al., 2009; Suzuki et al., 2003), carbon nanofibers (Joshi et al., 2010, 2012; Poudel et al., 2012), lampblack (Joshi et al., 2020; Lawaju & Joshi, 2022), and activated carbon (Chaudhary et al., 2024; Joshi et al., 2025; Kanjana et al., 2024) as alterative CE materials. These materials are much cheaper than Pt and possess catalytic property which is comparable to that of Pt. Among the carbonaceous CE materials mentioned above, the synthesis process of activated carbons derived from biomass is considered as eco-friendly one. Chaudhary et al. (Chaudhary et al., 2024) employed three types of carbons prepared from waste jute sacks as a CE material of DSSCs and reported a maximum power conversion efficiency (PCE) of 3.35% compared with 3.39% from a Pt-based DSSC. Recently, Joshi et al. (Joshi et al., 2025) have reported the DSSCs with CEs prepared from activated carbon of Lokta paper. The PCE of these carbon-based DSSCs was 5.12% compared to 5.58% efficiency from a Pt-based reference solar cell. Although previous research reports have shown that plant-based activated carbons can be employed as a low-cost catalyst in DSSCs, the efficiencies of the activated carbon-based DSSCs are still lower than that of Pt-based DSSCs, hence, the photovoltaic performance of the biomass-based catalysts should be improved. One of the reasons for the inferior performance of the DSSCs with activated carbon is high resistivity of CEs. Imoto et al. reported that adding carbon black (CB) to activated carbon reduced the resistivity of the activated carbon-based CEs and improved PCE of the DSSCs compared with the PCE of DSSCs whose CEs were solely prepared with the activated carbon (Imoto et al., 2003). In this research, we report a mixture of Lokta paper-based activated carbon and graphite powder as an alternative CE material of DSSCs. The catalytic ability of the CE material was evaluated using the technique of electrochemical impedance spectroscopy (EIS). The charge transfer resistance (Rct) of the catalyst was 1.59 Ω-cm2 which indicates that the mixture of Lokta paper-based activated carbon and graphite powder can be an efficient catalyst for the reduction of tri-iodide ions used in DSSCs.

Experimental methods

Preparation of activated carbon from Lokta Paper

The activated carbon was prepared in two phases. Firstly, scrap paper of Lokta (precursor), collected from local handicraft industries in Bhaktapur, Nepal, was carbonized at 400ºC in an inert atmosphere of nitrogen (N_2). Secondly, the carbonized precursor was chemically activated with $ZnCl_2$ (Joshi et al., 2025). The ratio of the mass of the precursor to the activating agent was 1:1. The $ZnCl_2$ treated Lokta paper-based char was heated in a quartz tube furnace (Zhengzhou Protech Technology Co., LTD, China) at 800ºC in an inert atmosphere for about 4 hours. The activated carbon was ground and washed with 0.1 M HCl and distilled water subsequently. Finally, the activated carbon was dried and filtered (Chaudhary et al., 2024; Joshi et al., 2025).

Structural and elemental analysis

Structural analysis of the activated carbon was carried out by XRD using Bruker D2 PHASER diffractometer ($\lambda = 1.54$ Å). The surface morphology of the activated carbon and that of the mixture of the activated carbon and graphite were explored by SEM using FEI Helios Nanolab 400 Scanning Electron Microscope. Similarly, the elemental composition of the mixture was investigated with the SEM-EDS system.

Evaluation of catalytic property

Electrochemical impedance spectroscopy (EIS) is a widely used technique to evaluate the catalytic ability of a material for the reduction of tri-iodide ions used in DSSCs. Hence, the catalytic ability of the mixture of the activated carbon and graphite powder was evaluated by EIS of electrochemical dummy cells. Unlikely a working DSSC, an electrochemical dummy cell does not consist of photoelectrode and CE, rather it is prepared by assembling two identical CEs coated with the catalyst under investigation (Joshi et al., 2023). In order to prepare such symmetrical dummy cells used in this research, carbon paste was prepared by grinding the Lokta paper-based activated carbon and graphite powder (Graphite Fine Powder Pract, Central Drug House (P) Ltd., India) with carboxymethyl cellulose (CMC) in a mortar. The amount of the activated carbon and graphite was 80% and 20%, respectively. The CMC of ~2% concentration in water was used as the binder. The paste was coated onto FTO-glass substrates by the technique of doctor blading, and the carbon film was dried. Two identical electrodes, coated with the carbon film, were assembled with a piece of parafilm as a sealant/spacer. Next, the electrolyte containing iodide-tri-iodide ions (Iodolyte AN-50 purchased from Solaronix, Switzerland) was injected into the gap between the two electrodes of the symmetrical dummy cell (Joshi et al., 2023, 2025).

The EIS of the symmetrical electrochemical cells was conducted by applying an AC signal of 10 mV amplitude and frequencies ranging from 0.1 Hz to 100 kHz using Interface 1010 (Gamry Instrument, USA). The EIS was carried out without a DC bias voltage in the dark. The Nyquist plot obtained from the EIS was fitted with an equivalent circuit using Gamry Echem Analyst software to extract charge transfer resistance (R_{ct}) at the catalyst and electrolyte interface, and the catalytic ability of the mixture of the carbons was evaluated (Joshi et al., 2025).

Results and discussion

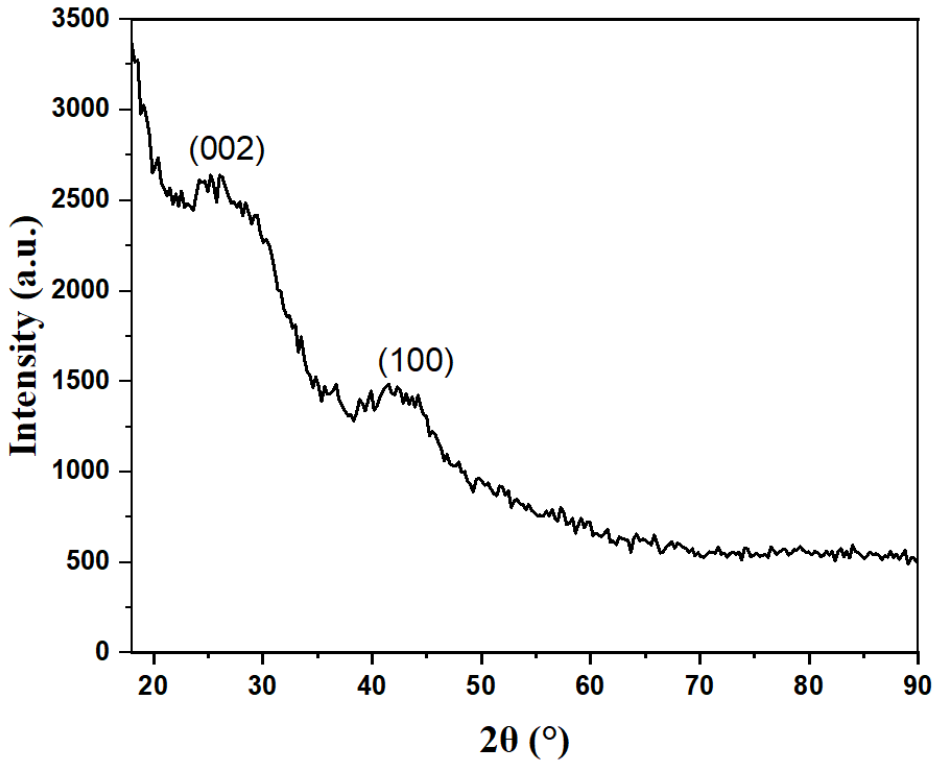

Figure 1: XRD of activated carbon

XRD of Lokta paper-based activated carbon

Figure 1 is the XRD pattern of the activated carbon derived from Lokta paper. Broad peaks centered near 2θ around ~25.22 to 26.02º and 2θ ~42.31º with significant background intensity indicate that the activated carbon contains graphite like carbon and amorphous carbon (Chaudhary et al., 2024; Joshi et al., 2025). Previous researchers have reported that such carbonaceous materials possess excellent catalytic ability for the reduction of tri-iodide ions used in DSSCs (Chaudhary et al., 2024; Joshi et al., 2010, 2025).

SEM images

Figure 2 shows SEM images of the Lokta-based activated carbon. Figure 2a is the SEM image at 10,000 times magnification. It shows that the activated carbon of Lokta comprises cylindrical and granular particles. The cylindrical shaped particles are several microns long, whereas the granular shaped carbons are more fragmented into micron to submicron sized particles. A significant number of micron sized gaps or voids among the particles are also seen in the SEM image. Figure 2b is the SEM image at 25,000 times magnification. It reveals that the surface of carbon particles is porous and rough.

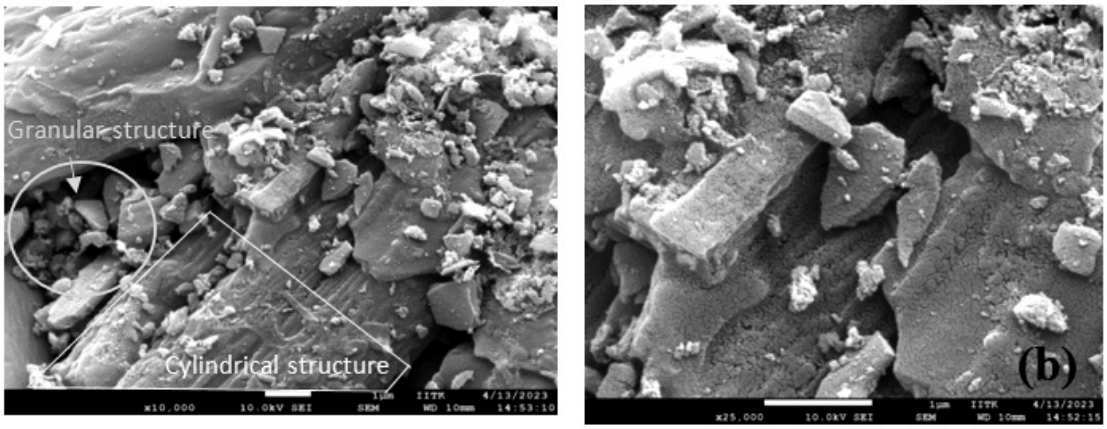

Figure 2: SEM image of activated carbon of Lokta paper at magnification of (a) 10,000 times and (b) 25,000 times.

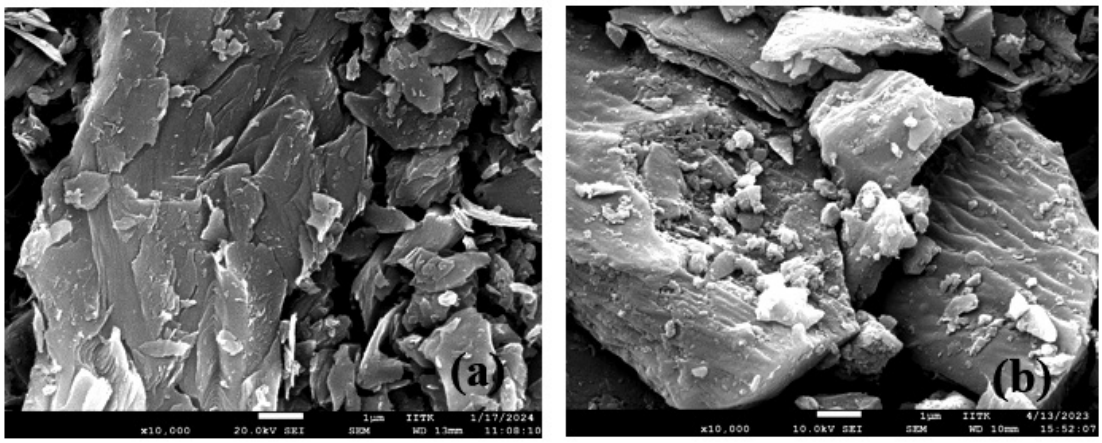

Figure 3: SEM image of (a) graphite at magnification of 10,000 times and (b) mixture of activated carbon (80%) and graphite (20%) at magnification of 10,000 times.

Figure 3a is a SEM image of the graphite at 10,000 times magnification. Figure 3b is a SEM image of the mixture of the carbons at 10,000 times magnification, and it shows two different types of materials. One of them is boulder shaped particles having dimensions of several microns. These particles are the dominant ones. The surface of these particles shows wrinkles and rough morphology. These features are analogous to those seen in the activated carbon. Hence, these are the particles of the activated carbon. The other type of material is seen as a bundle of thin sheets (as seen in the top part of the SEM image). The shape of this material is similar to that seen in Figure 3a; hence, the material of thin sheets is graphite. The SEM image also reveals that the graphite sheets are trapped among the boulder shaped particles. As graphite is highly conducting carbon, it may enhance the electrical conductivity of the mixture.

Elemental analysis of the mixture of activated carbon and graphite

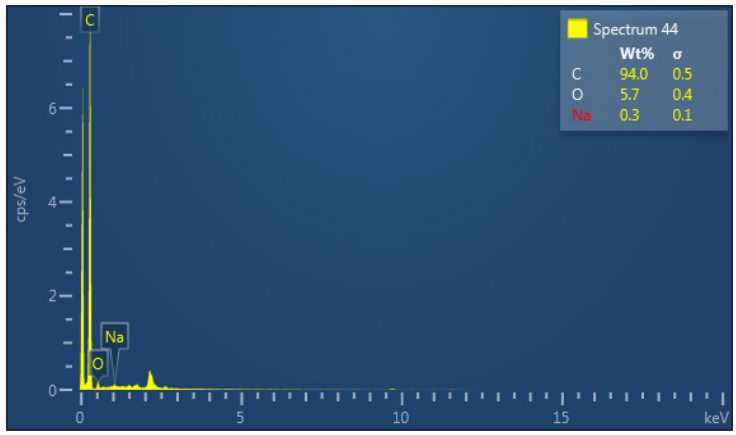

Figure 4: Typical EDS spectrum of the mixture of Lokta-based activated carbon and graphite powder.

In order to investigate the elemental composition of the mixture of Lokta-based activated carbon and graphite, EDS measurements were carried out at different points in the sample. Figure 4 shows a typical EDS spectrum of the mixture of the carbons. Various elements and their average approximate amounts detected in the sample were C (94.83%), O (4.73%), Na (0.08%), Cl (0.13%), Al (0.10%), S (0.08%), and Si (0.08%) indicating carbon as the major element in the sample.

Electrochemical impedance spectroscopy

Figure 5a shows the Nyquist plot of the electrochemical symmetrical dummy cell prepared with the mixture of Lokta-based activated carbon and graphite. Figure 5b is the equivalent circuit (Hauch & Georg, 2001) used to fit the Nyquist plot. In the equivalent circuit, R_s is the series resistance which is mainly the sum of ohmic resistance of the electrodes and electrolyte of the dummy cell. Similarly, R_{ct} is the charge transfer resistance at the catalyst-electrolyte interface. Another parameter, constant phase element (CPE), is a double-layer capacitor formed at the electrode-electrolyte interface. The other

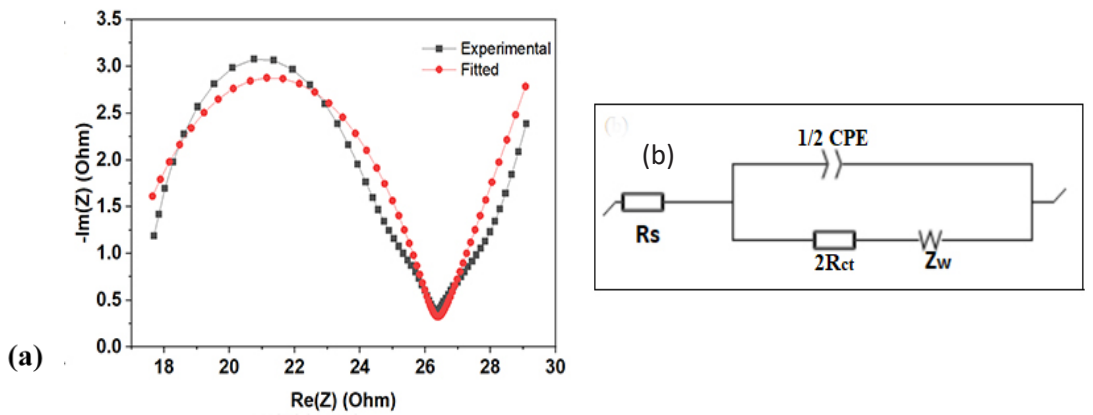

Figure 5: (a) Nyquist plot of electrochemical symmetrical dummy cell prepared with the mixture of Lokta-based activated carbon and graphite, (b) Equivalent circuit used to fit the Nyquist plot.

parameter Z_W is the Nernst diffusion impedance, which is related to the diffusion of ions in the electrolyte (Hauch & Georg, 2001; Joshi et al., 2010, 2023, 2025; Sun et al., 2018). Out of the circuit elements mentioned above, R_{ct} is directly related to the transfer of charge from the CE to the electrolyte or the reduction of the tri-iodide ions into iodide ions (Joshi et al., 2012, 2025). The value of $2R_{ct}$ (for two electrodes or the entire symmetrical cell) determined from the curve fitting of the Nyquist plot is 10.06 Ω (3.19 Ω-cm^2). The numerical value of this parameter indicates the ability of the catalyst for the reduction of tri-iodide ions into iodide ions. Generally, a smaller value of R_{ct} indicates better catalytic ability of the catalyst (Joshi et al., 2025). According to Hauch and Georg, the CE with R_{ct} of 10 Ω-cm^2 or less can be used in the fabrication of a DSSC (Hauch & Georg, 2001). In this work, R_{ct} is ~1.59 Ω-cm^2 (for single electrode), which is much smaller than the threshold value suggested by Haunch and Georg (Hauch & Georg, 2001). This indicates that the mixture of the activated carbon derived from Lokta paper and graphite powder can be an efficient catalyst for the reduction of tri-iodide ions in DSSCs.

Conclusions

The activated carbon, derived from Lokta paper and carbonized in two steps, was synthesized and characterized for the preparation of efficient CEs of DSSCs. The SEM image showed rough and porous surface morphology of the activated carbon, and XRD disclosed the existence of graphitic carbon and amorphous carbon in the sample. Aiming to enhance electrical conductivity, the activated carbon was mixed with graphite powder. The charge transfer resistance (R_{ct}) at the catalyst-electrolyte interface of the symmetrical dummy cell with the mixture of the activated carbon and graphite was 1.59 Ω-cm^2. Hence, the mixture of the activated carbon and graphite can be an efficient, low-cost, and eco-friendly non-platinum-based CE material for DSSCs.

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