



Multifunctional ammonium fuel cell using compost as a novel electro-catalyst



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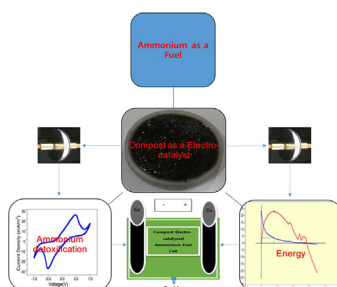
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HIGHLIGHTS

- Compost directly act as a abundant and cheapest electro-catalyst.
- Electro-catalytic activity leads to ammonium oxidation and generation of energy.
- Fuel cell is made by variation in electrodes, compost and ammonium concentration.
- Individual as well as joint physio-chemical/microbial action is possible.
- This work elucidates the development of multifunctional fuel cell.

GRAPHICAL ABSTRACT



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ABSTRACT

Due to acute ammonium toxicity, it is always desirable to find a cheaper and abundant electro-catalyst other than platinum, iridium oxide, boron diamond etc with a high selectivity and negligible de-activation for its oxidation. Also ammonium is not known for electricity generation except biological nitrification process. So this paper elucidates the studies of compost as a novel electro-catalyst in a ammonium fuel cell configuration. These studies are done by varying type of electrodes & compost as well as ammonium concentration. Bi-polar cyclic voltammetry, electrochemical impedance spectroscopy, temperature dependence, cyclic stability and chronoamperometry techniques are used to study compost. Cow dung based compost is found to show the best electro-catalytic activity. IV measurements are conducted to study power generation in tune with the electro-catalytic activity. Finally, polarization and sustainability measurements are done on a comparatively larger fuel cell to check the size scalability. The results shows that the maximum power density is 108 mW/m^2 and this multifunctional device can be fueled after every 12 h for continuous operation and with negligible de-activation of electro-catalyst. These studies opens a window for doing further advanced research in compost triggered electro-catalysis to make multifunctional fuel cell devices for solving environmental and energy issues together.

1. Introduction

The world today is facing a major problem of eutrophication and energy crisis at the same time so the development of multifunctional

hybrid devices is the need of the hour which can serve the dual purpose of environmental cleaning as well as generating efficient, sustainable and cost effective electricity. The electrochemical method has been shown for ammonia and ammonium removal from wastewater, landfill

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leachate, saline wastewater etc by using different electro-catalyst materials like boron-doped diamond (BDD), Platinum and IrO_2 etc. These electro-catalysts are associated with lot of problems for ammonia/ammonium oxidation applications such as high cost, limited selectivity and de-activation problem etc [1–6]. Other than electro-chemical method, there is another method in which micro-organisms can also degrade ammonia/ammonium using nitrification concept but it is a very slow process and takes longer time [7–12]. It is well known that soil based bio fuel cells can harness the energy from organic matter degradation by anaerobic bacteria or enzyme based redox reactions respectively where bacteria and enzymes act as a bio-electro-catalyst but nothing has been reported using soil/compost directly as an electro-catalyst without considering any direct role of bacteria and enzyme [13–20]. Ammonium chloride was known only as a very good ionic conductor until it shows the unusual ammonium oxidation behavior in aerobic/anaerobic conditions yielding minute electricity with the help of a specific bacteria *Nitrosomonas europaea* [11]. There is always a need for a new and cheaper electro-catalyst to achieve the goal of ammonium detoxification from ammonium rich waste waters. Electricity generation is also desirable as the world today is facing a major problem of energy crisis and the development of new type of renewable energy sources is the need of the hour [21]. Different type of soils/composts are abundant in nature, like both normal soil based compost and cow dung based compost, which are full of electronic and ionic abundance in nature due to the action of both aerobic and anaerobic bacteria in it, but due to their complex nature, this type of work is never motivated [19,20]. From the survey, it is clear, that nothing is reported on the use of soil/compost directly as an electro-catalyst for ammonium oxidation and production of electricity except costlier electro-catalysts or bacterial nitrification/denitrification. In this work compost based multifunctional device was designed for simultaneous ammonium detoxification and power generation based on the studies of compost directly as an electro-catalyst. To achieve this goal in a device mode, studies were done by varying type of electrodes, type of compost and ammonium concentration. Graphical view showing the vision of this paper is shown in supplementary information as Fig. 1. These studies lay a foundation for making next generation multi-functional devices for detoxification of ammonium from the environment and energy generation together by using cheaper compost as an electro-catalyst with high selectivity and negligible de-activation.

2. Experimental

2.1. Sample preparation

Initially both Cu/Cu and G/G combination of electrodes with a zero ammonium oxidation ability were chosen for the study of electro-catalytic activity [1–6]. Two types of composts supplied by Seung Jin Fertilizers Pvt Limited Seoul were used as the electro-catalyst. The main difference between them is that one was simple carbon rich soil based compost made from plant leaves and other one was nitrogen rich cow dung based compost made from cow dung manure. The surface texture of both composts was also studied by SEM measurements to see the difference in the surface texture which is shown in supplementary information as Fig. S2(a&b). Coin cell CR2032 normally used for Li Ion batteries [23–25] was used for the present studies. The coin cell was assembled in an open air using the Cu or a G foil disk (15 mm) as the working electrode and 3 g of compost in it, and again Cu or a G foil as the counter electrode. For electrode selection, some basic studies on electro-catalytic activity of both type of composts were done using distilled water (D.W) and 0.1 mg/ml of ammonium chloride. In comparison to G/G electrodes, Cu/Cu electrodes shows better electro-catalytic activity using compost as an electro-catalyst which is shown in supplementary information as Fig. S3 & S4. As compost is a bio-media so the earlier claim for highest bio-electrochemical activity in case of Cu/Cu electrodes seems to be justified [22]. Based on the preliminary

investigation on 0.1 g/ml of NH_4Cl , Cu/Cu electrodes were used for the whole electro-catalytic studies. A 0.1 g/ml, 0.2 g/ml and 0.3 g/ml of ammonium chloride dissolved in distilled water was used as the testing media for studying the effect of ammonium chloride concentration. The dimensions of the coin cell used in the above studies is as follows. Thickness of the cell = 0.20 cm, diameter 0.32 cm, Surface Area of the cell = 3.14 cm^2 . Similar type of coin cell is used for the basic IV measurements to optimize various parameters. After optimization of the similar coin cell for the best electrode conditions, a multifunctional fuel cell for real application was designed using Cu/Cu electrodes having areal electrode density of 17.5 cm^2 with 50 g of compost in it. A smaller hole at the bottom of the fuel cell was used to collect and remove the unwanted residue in it. Coding of the samples was done to avoid any confusion. For a soil based compost, code RSS was used for distilled water based sample and code SS was used for ammonium chloride based samples. For a cow dung compost, code RCSS was used for distilled water based sample and code CSS was used for ammonium chloride based samples. Even though the ammonium fuel cell was optimized considering compost as an overall electro-catalyst without considering the direct role of bacteria or enzyme, but to go in to the depth of mechanistic details, additional colony count studies using standard nutrient broth was done to see the growth of microbes on samples with/without ammonium as a feed. Colony counts provide information about bacterial counts or show those cells that were able of dividing and forming well defined colonies. Following procedure was followed for the colony count experiment which was used to demonstrate the cultivability of the soil bacterium. 0.1 g of soil (with or without ammonium) was first seeded into 9 mL peptone saline diluent (PSD) for 1 (hrs.) in incubation at ambient temperature. The inoculated PSD was further diluted with fresh PSD 1:9 for one time. Diluted soil suspensions (100 μL) were then directly plated onto the solid nutrient broth (NB) agar plates. The bacterial growth formed in between 0 and 48 (hrs.) incubation at 37°C were photographed, and viability rates of soil bacteria were thus examined.

2.2. Electrochemical characterization

The electrochemical properties of compost as an electro-catalyst were investigated using cyclic voltammetry (CV) in a bipolar mode, electrochemical impedance spectroscopy, temperature dependence studies, cyclic stability and chrono-amperometry. Electrochemically-active surface area (ESCA) of both the composts was also calculated to compare the electro-catalysts. The electro-catalytic performance of compost for ammonium oxidation in a coin cell layout for electro-catalytic bio-battery formation was investigated using MPG-2 16 channel battery cycler (Biologic Scientific Instruments, France) as shown in Fig. 1(a&b).

2.3. Electrical characterization

Two probe circuit using a Keithley high current source meter (Model-2420) interfaced with a computer in RS-232 mode was used to study the IV parameters of compost based electro-catalytic batteries which is shown in supplementary information as Fig. S5a. Initial IV measurements were done using Cu/Cu and G/G combination of electrodes in a same coin cell assembly that was used for electrochemical characterization as shown in Fig. 1(a). After optimization of the coin cell for the best IV conditions, a multifunctional fuel cell device for out of the lab application was designed using Cu/Cu electrodes as shown in Fig. S5b. Polarization curve was studied to see the maximum power obtained out of this compost based electro-catalytic battery. All the fuel cell related studies were done using natural reduction occurring at the cathode without any separator so that the vision of a multifunctional device can be achieved. The ammonium chloride concentration for the IV studies is fixed in tune with the best electro-catalytic activity achieved. To judge the relative amount of fuel which is left after fuel cell operation of 24 h for a one time addition of 50 ml of fuel at the

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