



# A modified decal method for preparing the membrane electrode assembly of proton exchange membrane fuel cells



Xiaolu Liang<sup>a,b</sup>, Guoshun Pan<sup>a,b,\*</sup>, Li Xu<sup>a,b</sup>, Jiashu Wang<sup>a</sup>

<sup>a</sup> State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China

<sup>b</sup> Shenzhen Key Laboratory of Micro-nano Manufacturing, Research Institute of Tsinghua University in Shenzhen, Shenzhen 518057, China

## HIGHLIGHTS

- The complete transfer is achieved by optimizing the catalyst inks' composition and a hot peeling technique.
- The thickness of catalyst layers (before transfer) is consistent.
- The effects of drying methods, transfer pressure, test temperatures are explored and discussed.
- The MEAs show better performance.

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## ABSTRACT

Decal transfer is an effective method for fabricating membrane electrode assemblies (MEAs), due to its low interfacial resistance and applicability for mass production. Here we introduce a modified decal method which makes it possible and convenient to achieve complete decal transferring. The composition of catalyst inks, drying process and transfer pressure are optimized in detail. During catalyst ink preparation, the viscosity is adjusted by altering the composition of the solvents to obtain catalyst layers (before transfer) with continuous thickness. In addition, MEAs whose catalyst layers (before transfer) were dried in four different ways are tested for application in a proton exchange membrane fuel cell. The transfer pressure is also optimized on the basis of the two previous conditions and the MEAs fabricated by this modified method show simplicity to achieve complete transfer in decal method, good repeatability and improvement in cell performance.

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## 1. Introduction

Proton exchange membrane fuel cells (PEMFCs) have attracted a great deal of attention due to several significant properties as a promising power sources for residential applications, vehicles, and portable electronic devices [1–3]. As the key component of PEMFC, the membrane electrode assembly, where both hydrogen oxidation and oxygen reduction reactions take place, is the major research area, especially studies on the fabrication. The decal method, in which catalyst inks are coated on decal substrates, dried and transferred to a membrane, is currently regarded as the most suitable approach for the commercial production of MEAs.

Most studies of the decal method usually focused on catalyst inks, drying methods, complete transfer and hot-pressing

conditions. Attention on catalyst inks can be classified as follows: (i) Coating methods, which are well known as spraying, doctor blading, and screen printing [1,4,5]. (ii) Dispersion solvents, whose volatility, viscosity and dielectric constant determine, to a great degree, the porosity of catalyst layers, the paintability of catalyst inks and the aggregation of Nafion ionomer [2,6–11]. (iii) Nafion-to-carbon ratio and solvent-to-carbon ratio. Plenty of studies focused on influence of Nafion content in catalyst layers on the resulting cell performance, and the reported Nafion loading varied from 20% to 50% along with Pt loading, weight percentage of Pt/C and preparation methods of MEAs [12–17]. As to the solvent-to-carbon ratio, both porosity of catalyst layers, which plays an important role on activation area and the mass transport rate, and residual organic solvents are research emphasis, especially the decal transfer method used [9,18]. (iv) Additives to improving water management by diminishing flooding in cathode catalysts layer, include pore formers, hydrophobic materials, oxide nanoparticles, and carbon materials which have fibrous morphology [3,19–24].

\* Corresponding author at: State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China. Tel.: +86 0755 26957243; fax: +86 0755 26957301.

E-mail addresses: [liangxiaolunihao@163.com](mailto:liangxiaolunihao@163.com) (X. Liang), [pangs@tsinghua.edu.cn](mailto:pangs@tsinghua.edu.cn) (G. Pan).

It is worth mentioning that, with the aid of optimized catalyst inks in the past several decades, improvement on performance of MEAs prepared by different methods is obvious to all [2,7,9–11]. In decal transfer method, catalyst inks are generally applied onto polymer films to form catalyst layers at first, by means of a doctor blade in general, and then transferred onto both sides of Nafion membrane by hot pressing. Numerous studies have concentrated on maximizing the transfer yield to cut loss of catalyst during the MEAs fabrication, since it is a critical and challenging target in decal method. Ha et al. [25,26] discussed the influence of decal selections on complete transfer, and the result proved that Telfon was far superior to Kapton. By employing a novel liquid nitrogen freezing method, transfer yield was also increased from 95.4% to 99.2%, presented by Cho et al. [27]. Moreover, after it was discovered that a thin dense Nafion ionomer “skin” formed during hot-pressing process, on account of strong hydrophobicity of the decal surface, Park and Cho et al. [25,28,29] made use of a carbon breaking layer to insure a high transfer ratio which was applied to decal before catalyst inks coating. Liquid treatment on electrolyte membrane was also proposed to achieve complete transfer which also made the process complicated [30]. It can be concluded that, it will be inspiring to achieve complete transfer via a convenient production process at the aid of optimizing catalyst inks coated on a suitable decal substrate.

In the past two decades, much attention has been focused on the high viscosity solvents used in catalyst inks, particularly glycerol [4,9,18,28,31,32]. Effects of addition, extraction and proportion of glycerol on cell performance were investigated in detail. However, as another organic solvent with high dielectric constant and viscosity, ethylene glycol which ever acts as the secondary pore former is less studied [33,34]. In this present research, deionized water, isopropyl alcohol and ethylene glycol are chosen as dispersion solvents. Influence of weight proportion of the three on viscosity is explored. On the basis of high-performing catalyst inks, a hot peeling technique is raised to achieve complete transfer without any auxiliary process. To optimize this modified decal method, effect of drying ways and transfer pressure on the single cell performance of MEAs is also discussed.

## 2. Experimental

### 2.1. Preparation of catalyst inks

The Pt/C catalyst (40 wt.% Pt content, Johnson Matthey), isopropyl alcohol, ethylene glycol, deionized water, and 5% Nafion solution (Dupont, Inc.) were mixed by an ultra turrax and in an ultrasonic bath at room temperature. The weight ratios of Pt/C catalyst, isopropyl alcohol, ethylene glycol and deionized water were varied. The viscosities of mixed solutions with different weight ratios were measured at 25 °C using a rotational rheometer (Malvern, Inc.). In all MEAs, the weight content of Nafion in catalyst layers is adjusted to 30% for all samples, and the Pt loading of cathode and anode is kept at 0.25 mg cm<sup>-2</sup> and 0.10 mg cm<sup>-2</sup> respectively.

### 2.2. MEA fabrication using the decal method

The prepared catalyst inks were coated onto decal substrates by an automatic film applicator. The fluorinated ethylene propylene (FEP) films were chosen as decal substrates. The blade gap of the automatic film applicator was set as 200 μm for the cathode and 100 μm for the anode. Meanwhile, the blade speed was kept at 38.6 mm/s for the cathode and 98.3 mm/s for the anode. After this, the decal substrates were dried in a vacuum oven or air drying oven at different temperatures (60 °C or 120 °C) for at least 4 h.

The Nafion HP membrane (Dupont, Inc.) was selected for the proton exchange membrane. The coated decal substrates were assembled by sandwiching the Nafion HP membrane and hot pressing under different pressures (4 MPa, 6 MPa, 8 MPa or 10 MPa) at 120 °C for 3 min. After the decal substrates were peeled off, the margin of MEAs was sealed by Kapton tape to shape a 25 cm<sup>2</sup> reaction area, which enhanced the MEAs' air tightness to a large degree. To obtain complete transfer, the whole hot peeling process was operated on the hot plate of the hot press machine whose temperature was still maintained at 120 °C. The two decal substrates were peeled off one by one in 15 s after the hot press machine stopped working. In contrast, decal substrates were also peeled off at room temperature, according to the traditional transfer process. Fig. 1 shows the whole MEA preparation process during this experiment.

### 2.3. Characterization

The morphological and elementary characteristics of catalyst layers and decal substrates were investigated using the FE-SEM (MIR3 XMH, Tescan, Ltd.). To obtain cross-sectional images, the decal substrates and MEAs were split by a knife and then stuck to a vertical surface. The pore size distribution of the catalyst layers was measured with a pore size analyzer (V-Sorb 2800P, Gold APP Instruments, Ltd.). The sealed MEAs were sandwiched between two carbon papers with microporous layers (GDS 3260, Avcarb, Ltd.) and then assembled with composite, one-serpentine flow field plates at a pressure of 20 kg cm<sup>-2</sup>. The performance of single-cells was tested using a Fuel Cell Test System (TEC-TS500, Top Energy, Ltd.) at 65 °C and ambient pressure. H<sub>2</sub> and air were supplied to the anode and cathode at flow rates of 700 ml min<sup>-1</sup> and 3000 ml min<sup>-1</sup>, respectively. The humidification temperature for both gases was maintained at 85 °C. The single-cells were activated at a constant voltage of 0.6 V for 4 h, then at 0.5 V for at least 20 h before characterization. The polarization curves were obtained by measuring the voltage using stepwise increments of current density, at intervals of 100 mA cm<sup>-2</sup>; each measurement lasted for 60 s. The EIS was tested at  $i = 1000$  mA cm<sup>-2</sup>, and the scanning frequency varied from 0.1 Hz to 10,000 Hz. The hydrogen humidification temperature, air humidification temperature and cell temperature were kept at 85 °C, 85 and 65 °C, respectively. The cell performance under different humidities was tested by altering the three above temperatures.

## 3. Results and discussions

### 3.1. Effect of the catalyst inks' composition on coating

In this experiment, paintability for catalyst inks is achieved when the carbon-to-solvent weight ratio (CSWR) is above 0.06. As the Nafion content is kept at 30 wt.%, the CSWR values only include isopropyl alcohol, ethylene glycol, and deionized water. To investigate further the effect of catalyst ink composition on coating, the viscosity of catalyst inks and cross-sections of catalyst layers before transfer are characterized. All the catalyst inks were applied to FEP films by the automatic film applicator. As samples #1, #2, and #3 in Table 1 show, the viscosity of catalyst inks increases as the CSWR changes from 0.06 to 0.12. Sample #4 substitutes ethylene glycol with the same weight of isopropyl alcohol that is in sample #2, whereas the opposite is the case for sample #5. Comparison of samples #2, #4 and #5 makes it clear that ethylene glycol increases the catalyst inks' viscosity, which influences their flow velocity when they are being coated onto decal substrates. It is evident that the viscosity of catalyst inks also can be adjusted through solvent selections, and paintability in the decal method can be improved by selecting high-viscosity solvents

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