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Technical Paper

High-rate roll-to-roll stack and lamination of multilayer structured membrane electrode assembly

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ABSTRACT

A membrane electrode assembly (MEA) is the heart of a proton exchange membrane (PEM) fuel cell stack. The presented roll-to-roll (R2R) system is initially developed for the mass production of MEA with high alignment precision, to avoid time-consuming manual manipulation. It is challenging to align large-area flexible multilayer structured membranes precisely. Two trilayer seal membranes are half-die-cut to structured films with various numbers of layers when their upper 2-layers are peeled off. Then the two structured trilayer seal films are laminated with skinless PEM to form composite PEM. In order to guarantee the manipulation precision and the registration of several multilayer structured films, the dark field illumination device and contact measurement device are developed to detect the micro-scale indentation on the transparent film. It can guarantee the discontinuous die-cutting process collaborates with the continuous R2R peeling and lamination processes, in a controllable production rhythm. The R2R technology is highly scalable for large area fuel cells.

1. Introduction

Fuel cells convert chemical energy directly into electrical energy with high-efficiency conversion, low emission of pollutants, high power density, and quiet operation [1,2].A membrane electrode assembly (MEA) is the heart of a proton exchange membrane (PEM) fuel cell stack [3]. Sheet components comprising a standard five-layer MEA that are cut to size to stack precisely: two gas diffusion layers (GDLs), PEM, and anode and cathode electro-catalyst layers [4]. Additionally, there are two other seal layers, anode and cathode layers. So far, fuel cells are usually prepared with the use of manual preparation with the low manufacturing efficiency and uniformity [5], because more than 9 flexible/brittle layers with full/half-cutted membranes have to be aligned together precisely. A challenge for the viability beyond laboratory depends on improving the efficiency and simplifying the preparation, such as the high-speed forming of fuel cell membranes as well as the seal lamination [6,7]. Hence, there is significant interest in this growing industry to continually improve PEM cell reliability and manufacturing efficiency, and also to reduce manufacturing costs.

Roll-to-roll (R2R) manufacturing system is an high-efficiency and low-cost approach, which has been proved in the field of solar cell [8,9], flexible display [10], RFID tags [11], flexible electronics [12]. R2R processing can continuously produce, and effectively save time of laying material. There are, however, still many challenges: (1) precise transmission and continuous lamination of ultra-thin flexible substrates; (2) synergy control and manipulation of picking-up, alignment, leveling, laminating and die-cutting; (3) high precision half-die-cutting of flexible sealing film; (4) online image capturing and micro-crack detection; (5) membrane cross-section changing in MEA process; (6) micro-tension control of ultrathin film; (7) uniformity of hot-pressing and temperature controlling; and (8) clamping of coarse-fragile-porous media.

This paper will present full-automatic R2R equipment of MEA, to upscale the manufacture from laboratory level to industrial level without any manual operation. The total R2R system contain peeling of trilayer film, half-cutting of bilayer film, seal lamination of two structured bilayer film. Detection technologies, including contact and noncontact manners, are presented to guarantee all these process carried out with the same rhythm and high position precision. The full-automatic R2R process can satisfy the requirements of commercialization and upscaled manufacture.

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2. Membrane electrode assembly and its compound technology

Fig. 1 shows the working principle of proton exchange membrane fuel cell (PEMFC) and the basic structures of MEA. The fuel is hydrogen and the charge carrier is the hydrogen ion (proton). At the anode, the hydrogen molecule is split into hydrogen ions (protons) and electrons. The hydrogen ions permeate across the electrolyte to the cathode while the electrons flow through an external circuit and produce electric power. Oxygen is supplied to the cathode and combines with the electrons and the hydrogen ions to produce water [13].A PEM is designed to conduct protons while being impermeable to gases such as oxygen or hydrogen. It is the essential function when incorporated into a MEA: separation of reactants and transport of protons. PEMFCs are permeable to protons when it is saturated with water, but it does not conduct electrons. GDLs are electrodes with a conjunction of a solid, liquid and gaseous interface, between which an electrical conducting catalyst supporting an electrochemical reaction. Oxygen and hydrogen react at the catalyst layer (CL), converting the chemical bond energy into electrical energy. Seal layers work as support in assembly of cells, meanwhile they can act as seal ring to avoid reactive gas bypassing the GDL. It is critically important to align large-area, multilayer flexible/brittle structured membrane precisely.

The framework of MEA production line is shown in Fig. 2. It can be divided into three relatively independent sections: R2R lamination, R2R catalyst coating, and R2R GDL overlaying. R2R lamination equipment is used to register and laminate ultra-thin seal membrane with PEM in high-efficiency and high-precision manners. Two kinds of processes have been developed to produce catalyst-coated MEA [14]: (1) CL is directly coated on PEM to form catalyst-coated membrane (CCM) which is then integrated with GDL; (2) CL is deposited onto GDL to generate gas diffusion electronic cathode which is then combined with PEM. The latter will produce huge waste of Pt catalyst. So we develop the CCM-based equipment, where the GDL integration is the last procedure, namely

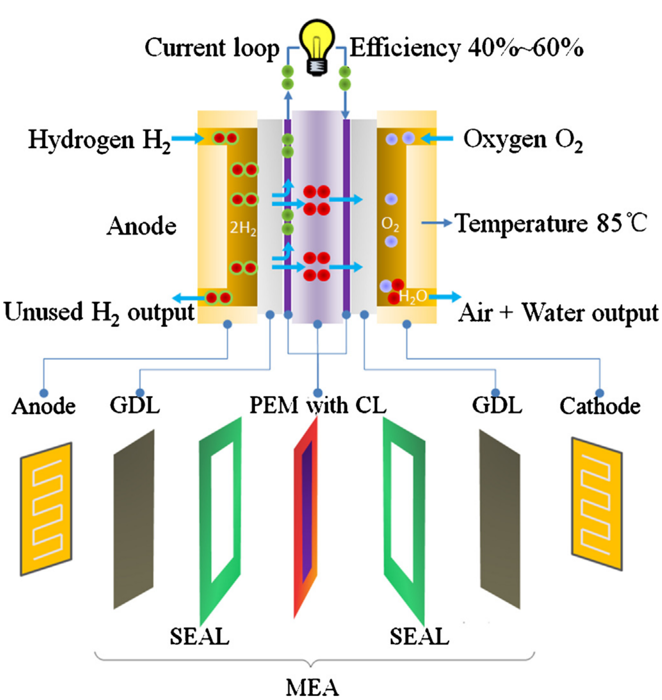

Fig. 1. Working principle of the proton exchange membrane fuel cell, and schematic diagram of MEA: PEM, anode catalyst layer, cathode catalyst layer, anode GDL, and cathode GDL.

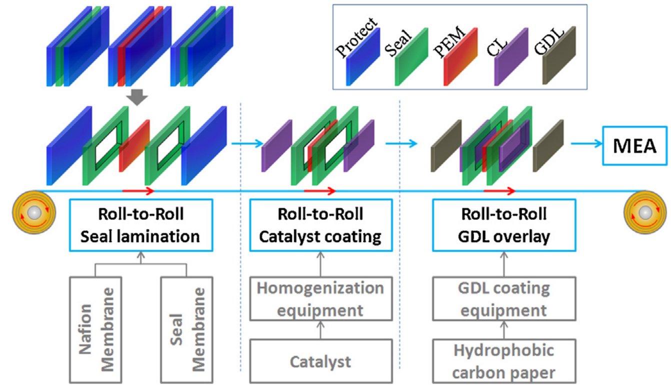

Fig. 2. The complete MEA production line of fuel cells based on R2R manufacturing system.

GDL overlaying. In the R2R lamination for CCM-based MEA manufacturing process, the encapsulation of MEA needs three trilayer membranes, which are two trilayer seal membranes and one trilayer Nafion membrane. The function of R2R lamination equipment is to peel 4 protective layers off, and to assemble a trilayer membrane (Seal-PEM-Seal) with upper and lower protective films into a 5-layer composite membrane. The seal is cut to form seal ring to protect GDL, but the two seal membranes need to be registered precisely. In the R2R catalyst coating section, the upper and lower protective films will be peeled off from the 5-layer composite membrane, and the CL will be coated on PEM through the Seal ring, which generates a new 5-layer composite membrane. In the last section, two GDL will be overlaid to assemble the 7-layer MEA.

3. Roll-to-roll stack and lamination system

3.1. Module design and layout

MEA needs multi-step processes, so the equipment contains the functions like die-cutting, waste film peeling, multiply laminations (initial and registered laminations), which are integrated together by R2R system. The seal lamination equipment is to half-cut the protective film of Nafion and peel the cut scrap of seal membrane off, then cold compress the seal-Nafion-seal to form compound membrane. The stack and lamination system is divided into six parts according to the functions, as shown in Fig. 3. (1) Preparing of upper seal membrane: the seal membrane is conveyed to die-cutting module to cut the upper two layers of trilayer seal membrane (Membrane I). (2) Peeling the upper protective film of Nafion membrane to produce 2-layer membrane, Membrane II, before the initial lamination. (3) Enabling initial lamination to integrate Membrane I with Membrane II to form 4-layer Membrane III. (4) Peeling the lower protective film of Membrane III to generate trilayer membrane, Membrane IV. (5) Preparing bottom seal membrane, similar with the upper one, to get Membrane V. (6) Registering lamination to combine Membrane IV with Membrane V to finish the final products. The registered lamination needs to be conducted while ensured the alignment of two cut squares of the upper and lower seal membranes.

Based on the design for MEA lamination process in Fig. 3, many modules should be appended, including: (1) tension control modules for the stable feeding of variable cross-section films; (2) guiding modules for films' lateral position control; (3) enough nip rollers for feeding, and idle rollers for layout arrangement; (4) various sensors for detections of tension, position, and images; (5) waste peeling modules for eliminating unnecessary rectangle seal sheets after die-cutting. The prototype of seal lamination equipment is shown as Fig. 4, which comprises unwind, die-cutting,