Journal of Zhejiang University SCIENCE
ISSN 1009-3095
http://www.zju.edu.cn/jzus
E-mail: jzus@zju.edu.cn

Influence of graphite particle size and its shape on performance of carbon composite bipolar plate*

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Received May 9, 2005; revision accepted July 26, 2005

Abstract: Bipolar plates for proton exchange membrane fuel cell (PEMFC) where polymer is used as binder and graphite is used as electric filler were prepared by means of compression molding technology. Study on the effects of graphite particle size and shape on the bipolar plate performance, such as electrical conductivity, strength, etc. showed that with decrease of graphite particle size, bulk electrical conductivity and thermometric conductivity decreased, but that flexural strength was enhanced. After spherical graphite occurrence in flake-like form, the flexural strength of the bipolar plate was enhanced, electrical conductivity increased but thermal conductivity decreased in direction paralleling pressure direction, and both electrical conductivity and thermometric conductivity reduced in direction perpendicular to pressure direction.

Key words: Spherical graphite, Fuel cell, Bipolar plate, Conductive composite materials
doi:10.1631/jzus.2005.A1080 **Document code:** A **CLC number:** TN911.5

INTRODUCTION

The proton exchange membrane fuel cell (PEMFC) is a very promising power source for residential and mobile applications due to its attractive features such as high power density, relatively low operating temperature, convenient fuel supply, long lifetime, etc. (Chalk *et al*., 2000; Lee and Mukergee, 1998). Despite these advantages, commercialization of PEMFC is delayed mainly due to the high fabrication cost. Bipolar plates, most commonly used among the PEMFC components, are the graphite plates with machined flow channels and the costs account for as much as 60% of the stack cost (Davies *et al*., 2000). So, for wide-spread commercialization of PEMFC, cost reduction of the bipolar plate is necessary. One approach to the cost reduction of PEMFC bipolar plates is to develop low manufacturing cost new materials such as polymer matrix composite. Cost models estimate that, with composites or metal alloys, the cost of bipolar plates would be only 15%~29% of the stack cost (Kirchain and Roth, 2002). In addition to low manufacturing cost, PEMFC bipolar plates require high surface and bulk electrical conductivity, sufficient mechanical strength, chemical stability in the PEMFC environment, gas tightness, and light weight (Borup and Vanderborgh, 1995). As a candidate to fulfill these requirements for PEMFC bipolar plate, carbon composite has been developed extensively (Scholta *et al*., 1999). However, most of the studies investigated the material properties of carbon composites or reported that, assembled into a single cell, composite bipolar plates exhibited performance comparable to that of metallic bipolar plates. There are few reported investigations on the effect of the kind of binder, its graphite particles size and shape on the performance of the bipolar plate.

Binder and electric powder are main components of polymer matrix composite bipolar plate, main factors affecting the performance of bipolar plate. As for effect of kind of binder, its content, and kind of

* Project (No. 2002AA323050) supported by the National Basic Research Program (863) of China

graphite and its content on bipolar plate performance, we have made significant progress (Zou *et al.*, 2004).

In this paper, the influence of graphite particles size and its granularity distribution on bipolar plate performance was investigated. In order to develop excellent performance composite bipolar plate to satisfy the need for higher powder cell stack, we also studied the effect of graphite shape on bipolar plate performance.

EXPERIMENTAL DETAILS

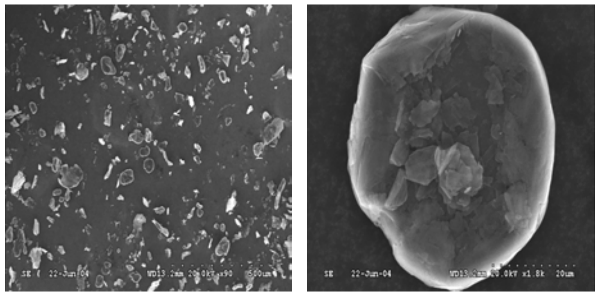
Materials

1. Binder

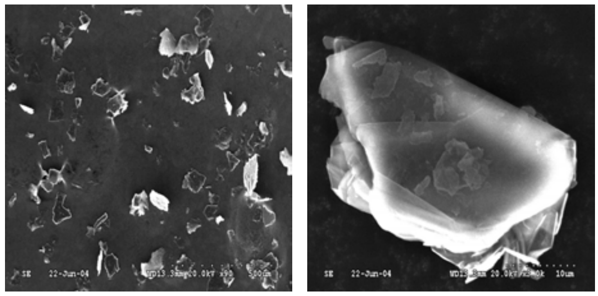
Polymer not only plays a binder role in the bipolar plate fabrication process, but is also one of the main factors affecting bipolar plate performance, such as electrical conductivity, flexural strength and so on. Polymer with polar atomic group in its molecular configuration, single electron or a pair of single electrons is easy to polarize or delocalize. And in this kind of polymer, electrical channels can easily be formed. Thus polymer with polar atomic group in its molecular configuration can appropriately be used as binder. Modified phenol resin with easy polar atomic group, such as –OH, and double bind, such atomic group and double bind are all beneficial for improving bipolar plate electrical conductivity. On the other hand, modified phenol resin has excellent affinity on the surface of the graphite, and is beneficial for increasing the mechanical performance of the bipolar plate. Therefore, modified phenol resin was used as binder in our experiment.

2. Electric powder

Natural flake-like graphite has better electrical performance than other kinds of graphite, so natural flake-like graphite was used as electric powder in our experiment. We chose three kinds of different particle size distribution natural flake-like graphite to use as electric powder with granularity distribution of 200~250 mesh, 250~325 mesh, and −325 mesh, respectively. A ball mill was used to grind the natural flake-like graphite into two kinds of different spherical natural graphite in granularity distribution of 250~325 mesh, −325 mesh, respectively. The electric powder morphologies were analyzed by scanning electron microscopy (SEM) (JEOL-Jsm-510, Japan). An SEM photograph is shown in Fig.1.



(a)



(b)

Fig.1 SEM micrograph of the spherical graphite (a) and flake-like graphite (b)

Sample preparation

Graphite powder with binder and solidified agent were put into negative pattern plates, under temperature of 160 °C and pressure of 4 MPa. Testing samples, 100 mm long, 40 mm wide, 10 mm thick, were prepared by hot press molding. The weight fraction of the electric powder was 84 wt% in all test samples. A 10 mm long, 3 mm wide, and 3 mm thick sample was prepared for determining the electrical conductivity and flexural strength. A 10 mm diameter and 2 mm thick sample used for determining thermometric conductivity was prepared by lathe.

RESULTS AND DISCUSSION

Electrical conductivity and thermometric conductivity in two directions (one parallel to pressure direction, the other perpendicular to pressure direction) and flexural strength of the sample were measured by sensitive micro-ohmmeter (HZ2522) meter, LJ500 type pull experimental machine, and laser thermometric conductivity instrument, respectively. Granularity distribution of the sample was analyzed by ultrasonic granularity analysis instrument.

Effect of granularity distribution on performance of bipolar plate

Results of tests on the effect of three kinds of different particles size flake-like graphite on the bipolar plate performance are summarized in Table 1.

In Table 1, "//" indicates electrical conductivity or thermometric conductivity parallel to pressure direction, "⊥" indicates electrical or thermometric conductivity perpendicular to pressure direction.

The results indicated that the electrical and thermometric conductivity of the samples had the same change trend with decreasing particles size and widening granularity distribution. The electrical mechanism of the composite materials involves mutual contact of the electric powder in the materials resulting in the formation of electrical channels. Natural flake-like graphite is an electric component, but binders are neither electric nor thermometric components, as the surface of graphite particles is coated with a non-conducting binder layer serving as a barrier preventing graphite particles from contacting each other to form electric networks. With decreasing particle size and widening granularity distribution, such barrier action notably decreases electrical and thermometric conductivity.

The particle size and granularity distribution of 200~250 mesh to −325 mesh flake-like graphite result in large scale increase of fine particle content and lead to decreasing size of the aperture between particles inside the matrix materials, increasing density, and increasing flexural strength.

Effect of shape of graphite particle on performance of bipolar plate

Graphite particles in composite materials orientate in direction perpendicular to pressure direction resulting in composite materials with relatively high electric conductivity in direction perpendicular to pressure direction. However, higher electric conductivity in direction paralleling pressure direction favors electric current transmission, so it is necessary to increase electrical performance in direction paralleling pressure direction. In order to increase electric performance in direction paralleling pressure direction, flake-like graphite is replaced by spherical graphite. The bipolar plate performances are listed in Table 2 after spherical graphite replacing flake-like graphite.

The results showed that, whether made of flake-like graphite or spherical graphite, the electrical and thermometric conductivity of the bipolar plate decreased along with decreasing the particles size and widening the granularity distribution, but because of the tiny-fine particles' reinforcing function, flexural strength of the sample made of –325 mesh graphite particles increased by 11%~13% over that of the sam-

Table 1 Effect of particle size distribution of natural graphite on the performance of the sample

Particle size (mm)	Direction	Bulk electrical conductivity (S/cm)	Thermal conductivity [W/(m·°C)]	Flexural strength (MPa)
200~250 mesh	//	78.8	21.00	27.5
	⊥	683.3	121.40	
250~325 mesh	//	76.0	20.30	35.0
	⊥	566.5	113.00	
−325 mesh	//	45.8	13.00	39.8
	⊥	420.3	105.47	

Table 2 Effect of graphite particle shape on properties of bipolar plate

Sample	Particle size	Direction	Bulk electrical conductivity (S/cm)	Thermometric conductivity [W/(m·°C)]	Flexural strength (MPa)
Spherical graphite	250~325 mesh	//	84.0	18.9	39.5
		⊥	387.3	71.9	
	−325 mesh	//	60.0	12.5	44.0
		⊥	326.9	61.4	
Flake-like graphite	250~325 mesh	//	76.0	20.3	35.0
		⊥	566.5	113.0	
	−325 mesh	//	45.8	13.0	39.8
		⊥	420.3	105.5	

ple made of 250~325 mesh graphite particles.

However, the bulk electrical conductivity in the direction paralleling pressure direction and flexural strength increased after spherical graphite was replaced by flake-like graphite. For example, flexural strength increased by more than 10%, electrical conductivity of the sample made of 250~325 mesh graphite particles increased by 11%, and that of sample made of –325 mesh graphite particles increased by 33%. This trend in which electrical conductivity increased in one direction, and in another direction weakened could be explained based on changing of graphite particles shapes. Fig.1 shows that flake-like graphite was altered in shape after milling by ball mill machine, that spherical and similar shape particles obviously increased in number, and that graphite particles electrical conductivity in the direction of perpendicular to pressure direction is weaker than that in the direction paralleling pressure direction. For PEMFC, higher electrical conductivity in the direction paralleling pressure direction is desired.

There are many factors which influence the strength of composite materials, but for composite materials including filler, whose shape is one of the main factors affecting composite materials strength, which in turn is mainly affected by the stress distribution around the particles inside composite materials and their strain. Compared with flake-like graphite, stress distribution around graphite particles is more homogeneous, and strain is smaller when spherical or similar spherical graphite are used as electric powder, so the flexural strength of bipolar plate increased.

On the other hand, the performance of bipolar plates relates to the shape of graphite particles, and to the content of spherical particles; the higher the content of spherical particles is, the more the electrical conductivity in the direction perpendicular to pressure direction. The flexural strength increases along with the increase of spherical graphite particles content. These test results are beneficial for developing excellent performance bipolar plates.

CONCLUSION

From the above study results, we conclude:

(1) The performance of the bipolar plate is related to graphite particle size and distribution. For flake-like graphite or spherical graphite, electrical and thermometric conductivity decrease with decreasing electrical particles size and widening distribution, but strength increases.

(2) After flake-like graphite is replaced by spherical graphite particles, both electrical conductivity (in the direction paralleling pressure direction) and flexural strength increase, but thermometric and electrical conductivity in the direction perpendicular to pressure direction reduce. For example, strength increases by more than 10%, electrical conductivity of the sample with 250~325 and –325 mesh graphite particles increases by 11% and 33% respectively.

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