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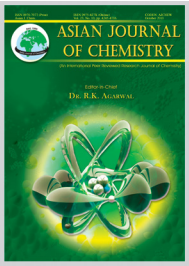

Crystallization Behaviour of MgB_2 Films Fabricated on Different Cathodes *via* Electrochemical Technique

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An electrochemical technique has been devised and settled to fabricate superconducting MgB_2 films in molten salts. $MgCl_2$, $Mg(BO_2)_2$, NaCl and KCl were used as electrolyte, graphite was used as anode and graphite and stainless steel were chosen as cathode, respectively. In the process of electrolysis, MgB_2 films were fabricated under Ar-gas flow. X-ray diffraction analysis was adopted to investigate the phase composition and crystallization property of the films. The results indicate that MgB_2 films have been fabricated successfully on different cathodes and the optimized electrolysis temperature are 620 ºC for graphite and 606 ºC for stainless steel. Furthermore, pulse power supply can enhance the content of MgB_2 phase.

Key Words: MgB_2 film, Electrochemistry, Crystallization property.

INTRODUCTION

Since the discovery of superconductivity in MgB_2, a lot of activities both in the application aspects and superconducting mechanism have been stimulated[1,2]. Up to date, the most common processing technique about MgB_2 cables preparation is based on powder-in-tube (PIT) method[3-6]. However, the low scalability and high processing cost of the technique to fabricate MgB_2 tapes or wires is undesirable and it is necessary to develop an alternative to powder-in-tube in order to make full use of the high potential of MgB_2 as a practical superconductor.

In recent, a processing technique of MgB_2-based superconducting products named the molten-salts electrochemical technique was reported[7-11]. The installations of the technique are very simple and the cost is also low and it is a way to prepare MgB_2 films with a thickness of several micrometers on cathode with various size and shapes. Moreover, films prepared by this method possess high enough superconducting characteristics ($H_{c2}(0) = 28$ T) to compete with those of powder-in-tube cables ($H_{c2}(0) = 18$-22 T), which make it possible to apply the technique as a promising alternative to powder-in-tube. However, the research of the technique is still in elementary stage. The technique has been applied only on few cathodes and the electrolytic conditions especially the electrolytic temperature are not fixed. Furthermore, the focus of the expriment is concentrated on the superconducting transport properties of MgB_2 films. In previous study[11], we prepared MgB_2 films on copper cathodes and investigated the influence of electrolyte temperature on the MgB_2 phase purity and superconducting transport properties. However, investigation on phase purity and crystallization property of MgB_2 films prepared on other cathodes is unsystematic.

In this paper, the crystallization behaviour of MgB_2 films fabricated on graphite and stainless steel cathode *via* the molten-salts electrochemical technique were investigated systematically. Electrolyte temperature dependence of phase purity was fixed and crystallization behaviour of MgB_2 phase on different cathode substrate was also investigated.

EXPERIMENTAL

Fig. 1 is a schematic diagram of a home-made electrolysis cell. A graphite crucible was used as anode as well as an electrolyte container. The electrolyte of the experiment was composed of $MgCl_2$, $Mg(BO_2)_2$, NaCl and KCl powders with a molar ratio of 10:2:5:5. In the process of preparation, raised the temperature to 400 ºC slowly and preserved the temperature for 2 h, under a dry Ar-gas flow. Then, heated to the electrolysis temperature, followed by waiting for 30 min, the electrolysis was stared by applying a constant DC voltage of 4V between the two electrodes for different electrolysis time. Furthermore, pulse power supply (voltage: 4V, frequency: 100 Hz, duty cycle: 50 %) was also adopted to compare the influence of power supply. After the electrolysis process, the cathode was

pulled out of the electrolyte but still in the furnace and began to cool. Took out of the samples when the temperature fell to room temperature and the solidified electrolyte sticking to the substrate was washed off with dry methanol using an ultrasonic washer. Experimental parameters for samples obtained under different electrolytic temperature on different cathodes are listed in Table-1. Phase compositions and crystal structures were characterized by XRD analysis (CuK_α radiation, scanning rate: 2º min).

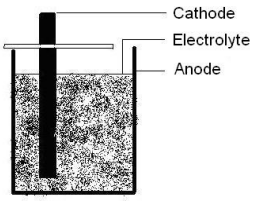

Fig. 1. Schematic diagram of an electrolysis cell

TABLE-1
EXPERIMENTAL PARAMETERS OF SAMPLES FABRICATED ON DIFFERENT CATHODES

Samples	Cathodes	Electric temperature (ºC)	Electric time (h)	Power supply
1#	Graphite	596	2	dc
2#	Graphite	610	2	dc
3#	Graphite	620	2	dc
4#	Graphite	638	2	dc
5#	Stainless steel	594	1	dc
6#	Stainless steel	598	1	dc
7#	Stainless steel	603	1	dc
8#	Stainless steel	598	1	Pulse

RESULTS AND DISCUSSION

Graphite cathode: As shown in Fig. 2, samples 1#-4# were prepared at 596, 610, 620 and 638 ºC for 2 h, respectively. (101) and (100) peaks of MgB_2 phase are occurred in samples 1#-3#, while there is only faint (101) peak in sample 4#. Therefore, 638 ºC is inappropriate temperature for the formation of MgB_2 phase. Moreover, the relative intensity of sample 3# is strongest in samples 1#-3#, which demonstrate that sample 3# possess higher content of MgB_2 phase. In addition, full width at half maximum (FWHM) of MgB_2 (101) peaks of samples 1#-3# is 0.21º, 0.22º, 0.19º, respectively, indicating an optimal crystalline perfection of MgB_2 phase in sample 3#. Therefore, the optimal electrolyte temperature is 620 ºC for samples prepared on graphite cathode. It should be noted that there are appearance of impurity peaks in all samples including sample 3#. The problem was also experienced by Abe group[7], which hints that it is difficult to get rid off the residual solidified electrolyte sticking to the substrate. The investigation to solve the challenging problems is in progress.

Stainless steel cathode: As shown in Fig. 3, samples 5#-7# were prepared at 594, 598 and 603 ºC for 1 h, respectively. There is no distinct diffraction peaks of MgB_2 phase in sample 7#, which may hint that the decomposition of MgB_2 phase at 603 ºC. Faint MgB_2 peaks occur in sample 5#, while diffraction peaks of MgB_4 and other impurities are strong. The reason may lie in the mobility of electrolyte is undesirable at 594 ºC, which may lead to the insufficiency for Mg element diffusion. Sample 6# possess higher content of MgB_2 phase, *i.e.*, sample 6# has a superior of phase purity of MgB_2. Therefore, the optimal electrolyte temperature is 598 ºC for samples prepared on stainless steel cathode.

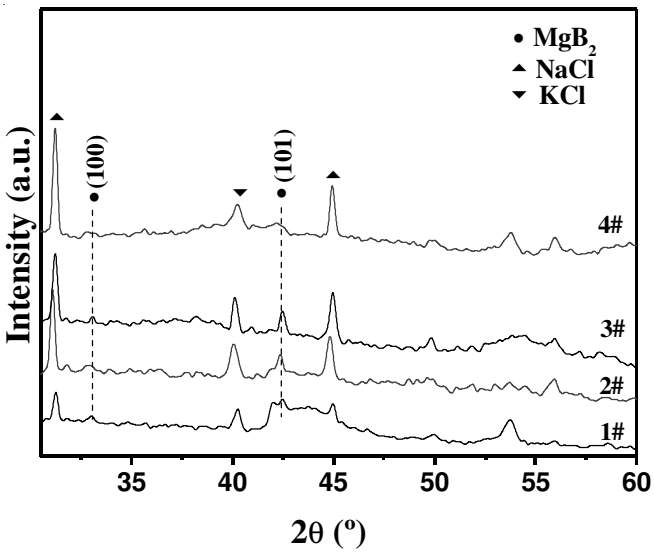

Fig. 2. XRD patterns of the samples prepared on graphite cathode and the electrolytic temperature for samples 1#-4# is 596 ºC, 610 ºC, 620 ºC, 638 ºC, respectively

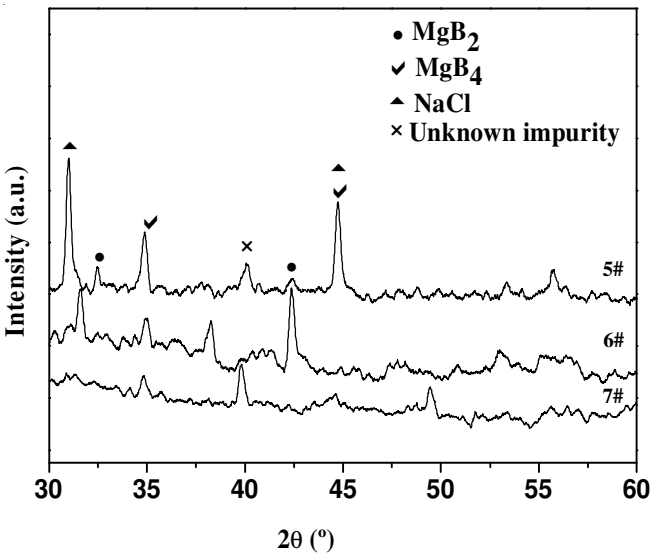

Fig.3. XRD patterns of the samples prepared on stainless steel cathode and the electrolytic temperature for samples 5#-7# is 594 ºC, 598 ºC and 603 ºC, respectively

In order to further investigate the influence of power supply mode on the phase purity and crystalline perfection of samples, pulse power supply was adopted to prepare sample 8# and the electrolyzed parameters were same as sample 6#. XRD patterns of samples 6# and 8# are shown in Fig. 4. Compared with sample 6#, content of MgB_2 phase increased in sample 8# and little MgB_4 phase and other impurity can be found. Furthermore, FWHM of MgB_2 (101) peak of sample 8# (0.52º) is much greater than that of sample 6# (0.32º), which hints a supervisor crystalline perfection. Therefore, pulse power supply can enhance the crystallization properties of MgB_2 phase. The reason may lie in the intermittent power

supply of pulse power supply can provide a sufficient diffusion duration for metal ions, which can largely maintain the ion concentration in molten salts. As a result, the crystallization property of MgB_2 phase is enhanced.

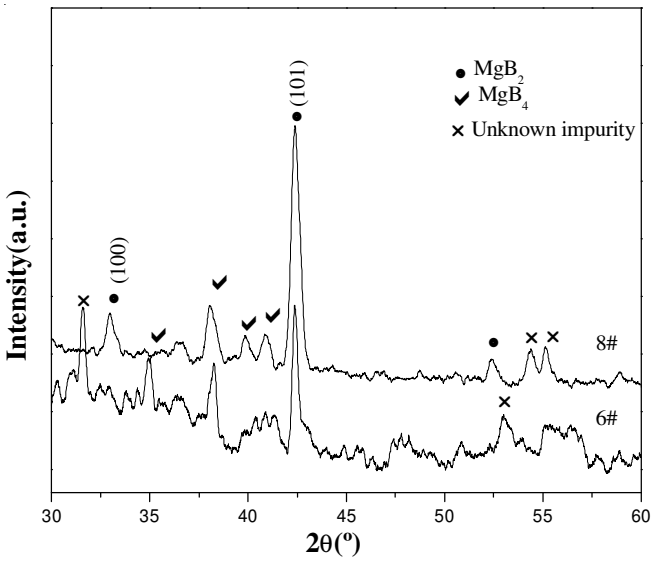

Fig. 4. XRD patterns of samples electrolyzed via common power (6#) and pulsed power (8#) on stainless steel cathode

Conclusion

MgB_2 films were fabricated successfully on different cathodes and the optimized electrolysis temperature are 620 ºC for graphite and 598 ºC for stainless steel; pulse power supply can largely maintain the ion concentration in molten salts and thereby enhance the crystallization properties of MgB_2 films.

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