# DEVELOPMENT OF LOW THERMAL EXPANSION GLAZE CORDIERITE BODY

# Wanna T. Saengchantara\*, Lada Punsukumtana, and Pranee Junlar

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# Abstract

A cordierite body with a low thermal expansion coefficient of  $1.5-4 - 10^{-6/\circ}$ C has excellent thermal shock resistance and can be used for manufacturing stove-top ceramic cookware. This study is aimed at developing a glaze for a cordierite body having a thermal expansion coefficient less than  $3.09 \times 10^{-6/\circ}$ C. The glazes in a Li<sub>2</sub>O-KNaO-CaO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system were calculated and prepared. The selected raw materials were mixed and ground in a ball mill. The samples were dip coated onto the cordierite body and fired at 1300°C with 2 h soaking time. After firing, the good visual crack-free samples were tested for crazing resistance by autoclave at 250 psi. The phases and thermal expansion coefficient were also analyzed by XRD and a dilatometer. The selected sample was applied to 7 inch diameter cordierite pots and these were tested for cooking on a gas stove-top or hot plate for more than 50 cycles. The results show that the glaze composed of virgilite and a zircon phase had a thermal expansion coefficient of  $2.61 \times 10^{-6/\circ}$ C, and was smooth and crack-free.

Keywords: Cordierite, cookware, low expansion, glaze

## Introduction

Cordierite, a magnesium aluminum silicate (2MgO•2Al<sub>2</sub>O<sub>3</sub>•5SiO<sub>2</sub>) from a mixture of talc, clay, and aluminum oxide, has low thermal expansion properties and excellent thermal shock resistance. Therefore, cordierite is used for parts requiring thermal shock resistance in order to endure the repeated cycles of rapid increases and decreases in temperatures, such as automobile catalytic converter supports, kiln furniture, and cookware (Bind, 1983).

The limitation of cordierite cookware production relates to the surface coating. It is

difficult to obtain a fully vitreous glaze due to the coefficient of thermal expansion (COE) of cordierite which has been generally known as  $1.5-4.0 \times 10^{-6/\circ}$ C. However, it is possible to glaze cordierite by introducing an appropriately formulated glaze which precipitates a low expansion phase in the vitreous matrix.

The primary crystal system used is lithia-alumina-silica (Li<sub>2</sub>O-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>) which has 2 low expansion crystals, beta eucryptite and beta spodumene. Cordierite in the magnesia-alumina-silica (MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>)

Department of Science Service, Ministry of Science and Technology, Bangkok, 10400, Thailand, Tel.: 0-2201-7034; Fax: 0-2201-7408; E-mail: wanna@dss.go.th

<sup>\*</sup> Corresponding author

system can also be used (Eppler and Eppler, 2000).

The objective of this work was to develop a low thermal expansion glaze having a smooth, hard, and crack-free surface for a cordierite body with the COE of  $3.09 \times 10^{-6/\circ}$ C and water absorption of 12%, and which could be used for producing cookware.

## **Materials and Methods**

The glaze of the  $Li_2O$ -KNaO-CaO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system used and the empirical formula is listed below. The mole of both Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> was varied as shown in Table 1. Ten wt% zircon was added to increase the opacity and hardness of the glaze (Hopper, 2001).

0.44 Li<sub>2</sub>O 0.68 – 0.88Al<sub>2</sub>O<sub>3</sub> 4.8 – 6.8 SiO<sub>2</sub> 0.12 KNaO 0.16 CaO 0.28 MgO

The selected raw materials were spodumene from CRU Co. Ltd., flux filler, CeraSil®, and Magneton from IMD Co., Ltd, Bangkok, Thailand, dolomite, talc, and zircon from Cernic International Co., Ltd., Nakhon Pathom, Thailand, and alumina A31 from Niso Co., Ltd. The batch compositions in weight% were in the following ranges: 30-41% spodumene, 10-14% flux filler, 6-8% dolomite, 4-5% talc, 6-14% Magneton, 2-3% alumina A31, 20-42% Cerasil®, and 10% zircon.

The batches were finely ground in a ball mill and sieved through 140 mesh. The prepared glazes were applied on the biscuit cordierite body and fired at 1300°C at the rate of 150°C /h and soaked for 2 h. After firing, the glazed samples were determined by visual examination and the good, visually crackfree samples were evaluated. The crazing resistance by autoclave at 250 psi (following ASTM C424-80; (ASTM, 1989)), phase analysis by x-ray diffraction (Bruker D8-Advance, Bruker Corp., Billerica, MA, USA), and COE analysis by dilatometer (Netzsch DIL402PC, Netzsch-Geratebau GmbH, Selb, Germany) were used to analyze the samples. Then the selected sample was applied to 7-inch diameter cordierite pots. The 20 pots were thermally shock tested with a boiling test on a hot plate and quenching in cool water, then the crack lines were examined. The thermal shock tests were repeated for 5 cycles. When no crazing was found in a sample, the sample was then thermally shock tested on a gas stove-top for 5 cycles. Finally the pots were tested by cooking on a hot plate or a gas stove-top for more than 50 cycles.

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Table 1 Mole of Al<sub>2</sub>O<sub>2</sub> and SiO<sub>2</sub>

Glaze sample No.	Mole of Al <sub>2</sub> O <sub>3</sub>	Mole of SiO <sub>2</sub>
LG2.1	0.68	4.8
LG2.2	0.78	4.8
LG2.3	0.88	4.8
LG2.4	0.68	5.8
LG2.5	0.78	5.8
LG2.6	0.88	5.8
LG2.7	0.68	6.8
LG2.8	0.78	6.8
LG2.9	0.88	6.8

#### **Results and Discussion**

The characteristics of the glaze samples by visual examination are shown in Table 2. The results showed that most of the glaze samples were opaque, glossy, semi-glossy, or crazed except for glaze sample L2.9. After the crazing resistance test of glaze sample L2.9 up to 250 psi, the tested sample was found to have no cracks, as shown in Table 3. The COE of the cordierite body and glaze sample L2.9 are shown in Figure 1. The results showed that the COE of the glaze,  $2.61 \times 10^{-6/\circ}$ C, was lower than the COE of the cordierite body,  $3.09 \times 10^{-6/\circ}$ C. That is the reason why the glaze sample used on the cordierite body with water absorption of 12% had no cracks after the crazing resistance test (Parmalee, 1973). The crack-free surface is highly recommended for cookware because cracks attract bacteria and are vulnerable to degrading when exposed to strong basic substances such as dishwasher detergents as well as the acids that are present in food (Britt, 2004). The x-ray pattern in Figure 2 confirmed that the glaze sample fired at 1300°C was composed of 2 crystal phases, virgilite and zircon. Both of the crystals have low thermal expansion (Kingery *et al.*, 1991; Soares *et al.*, 2011).

The results for the cordierite pots glazed with the L2.9 sample and tested on a hot plate and a gas stove showed that they were found to be crack-free, as shown in Figure 3. Figure 4 shows the cooking test of a cordierite pot glazed with glaze sample L2.9. The glaze was also crack-free after the cooking test for 50 cycles.

#### Conclusions

The glaze in the  $Li_2O$ -KNaO-CaO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system with the empirical formula

Table 2.	The characteristics o	f tested glaze	samples on co	ordierite body fired at	1300°C
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	Glaze appearance			
Glaze sample No.	Transparent-Opaque	Gloss-Matte	Crazed	
LG2.1	Opaque	Gloss	$\checkmark$	
LG2.2	Opaque	Gloss	$\checkmark$	
LG2.3	Opaque	Gloss	$\checkmark$	
LG2.4	Opaque	Semi gloss	$\checkmark$	
LG2.5	Opaque	Semi gloss	$\checkmark$	
LG2.6	Opaque	Semi gloss	$\checkmark$	
LG2.7	Opaque	Semi gloss	$\checkmark$	
LG2.8	Opaque	Semi gloss	$\checkmark$	
LG2.9	Opaque	Semi gloss	Х	

Table 3. Crazing resistance test of glaze sample L2.9 on cordierite body fired at 1300°C

Glaze sample No.	Cordierite body fired at 1300°C				
	Autoclave pressure, psi				
_	50	100	150	200	250
LG2.9	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$



Figure 1. COE of cordierite body used and the glaze sample no. L2.9



Figure 2. XRD pattern of the glaze sample L2.9 fired at 1300°C



Figure 3. The test on a hot plate and a gas stove-top of the cordierite pot glazed with L2.9

of 0.44 Li<sub>2</sub>O 0.12 KNaO 0.16 CaO 0.28 MgO 0.88Al<sub>2</sub>O<sub>3</sub> 6.8 SiO<sub>2</sub> after firing at 1300°C had a smooth surface, was crack-free, and had the COE of  $2.61 \times 10^{-6}$ /°C. It could be applied to the cordierite body with the COE of  $3.09 \times 10^{-6}$ /°C and was proven to have thermal shock resistance and to be suitable for producing cookware.

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Figure 4. The cooking test of the cordierite pot glazed with L2.9