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Influence of mineralogical composition on the properties of lightweight aggregate

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ABSTRACT

When sintering artificial lightweight aggregate, rapid cooling will generate micro-cracks. The paper investigates the effect of MgO contents on the physical, chemical and microstructural properties of artificial lightweight aggregate prepared under natural cooling in the furnace (SC) and rapid cooling outside the furnace (RC). Results indicate that cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) starts forming in lightweight aggregate when MgO content reaches 2%, accompanied by a reduction in mullite. Cordierite has a better thermal-shock resistance and its presence minimizes the potential for micro-crack formation, thus decreasing water absorption under rapid cooling conditions for the 2% MgO system.

1. Introduction

The concept of high performance lightweight aggregate (HPLWA) was first proposed in the early 1990s in Japan with the aim to improve the pumpability of lightweight aggregate concrete (LWAC). However, there exists no consensus regarding the definition of HPLWA among researchers in China and abroad. Generally speaking, HPLWA is high in strength and low in water absorption both under conventional and pressure saturation, and is also well graded [1,2]. Ideal lightweight aggregate pellets should have [3]:

(1) a strong but low density, porous, sintered ceramic core;
(2) a dense continuous surface layer to inhibit ingress of water that should be pozzolanic to produce a strong aggregate–cement bond in concrete;
(3) a near-spherical shape to improve fresh concrete properties.

Currently, rapid cooling is the most common way of cooling in the preparation of LWA; meanwhile, micro-cracks are also commonly generated on or underneath its surface, undermining its microstructure and thus its mechanical and physical properties. Accordingly, much research has been undertaken on raw material composition, sintering regime, etc. to solve this hazard [4–9], but little, if any, on the mineralogical composition and its concomitant effect on macro properties of LWA. The mineral composition of the various lightweight aggregates varies somewhat depending on raw materials and manufacturing procedures. The main minerals identified for the aggregates are quartz, anorthite, ringwoodite, hematite, and mullite [10], with mullite being the main phase responsible for the strength of LWA [11–13].

Cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) is characterized by a low coefficient of thermal expansion (2×10^{-6}–$6\times10^{-6}\ ^{\circ}C^{-1}$) and an excellent thermal-shock resistance [14] and is often introduced into a multi-phase composite system to improve its thermal stability [15–17]. In the paper, MgO is introduced to form the cordierite phase in LWA pellets and the resultant effect on their properties is investigated.

2. Materials and experimental program

2.1. Materials

LWA pellets were sintered from a combination of fly ash, expanded shale, Kaolin and quartz powder. Basic magnesium carbonate ($3MgCO_3{\cdot}Mg(OH)_2{\cdot}3H_2O$) was added to introduce MgO. The chemical properties of the raw materials are indicated in Table 1.

2.2. Mixture proportions

The recipe of the control mixture was listed in Table 2 as MS-0 (MR-0), where MS indicates natural cooling in the furnace, MR indicates rapid cooling. The mixture consisted of 33.92 wt.% fly ash, 27.11 wt.% kaolin, 9.53 wt.% shale and 29.44 wt.% quartz. Different contents of basic magnesium carbonate were added to MS-0 (MR-0) for varying the contents of MgO. MS-2 (MR-2), MS-4 (MR-4) and MS-6 (MR-6) were obtained, the MgO contents by weight of which were 2%, 4% and 6%, respectively.

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Table 1
Chemical composition of raw materials, wt.%.

	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	SO_3	P_2O_5
Fly ash	47.89	28.82	6.64	3.35	1.06	2.15	0.56	2.18	1.27	0.34
Expanded shale	74.50	13.52	1.21	1.00	0.29	4.50	4.47	0.18	0.041	0.070
Kaolin	48.61	36.45	0.75	0.014	0.11	0.88	–	0.29	0.16	0.039
Quartz powder	98.95	0.64	0.028	0.015	–	0.10	–	–	–	–

Table 2
Chemical composition of the control mixture, wt.%.

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	TiO_2	$K_2O + Na_2O$	LOI
65.65	21.14	2.58	1.24	0.42	0.84	2.01	5.27

2.3. Preparation of LWA pellets

LWA pellets were prepared according to Fig. 1. The granulation of Φ12 × 16 mm cylindrical pellets was by plastic extrusion. A Si–Mo bar furnace (RJX-10-16) with a 400 × 200 × 150 mm furnace cabinet was used for sintering and the pellets were first pre-heated at 500 °C and then heated at a constant rate of 5 °C/min to 1200 °C, held for a dwell time of 20 min and finally cooled to room temperature. Two cooling processes were employed: natural cooling in the furnace (SC) and rapid cooling outside the furnace (RC).

2.4. Physical and mechanical testing

The compressive strength of LWA pellets was determined. Prior to testing, the upper and lower surfaces of the cylindrical sample pellets were polished by a metallographic polishing machine, and then coated with a layer of epoxy resin. After hardening, the two surfaces were again polished with abrasive paper. Finally, pellets were loaded to fracture by a MTS Ceramic Test System.

The dry density and water absorption of LWA pellets were calculated by measuring the dry mass m_{dry}, mass immersed in water m_{imm} (measured by hydrostatic balance), 24 h saturated-surface-dry mass immersed in water for 24 h $m_{24\,h}$ and vacuum saturated mass m_{vac} Saturated-surface-dry (SSD) [18] is defined as the condition in which the permeable pores of aggregate particle are filled with water to the extent achieved by submerging in water for the prescribed period of time, but without free water on the surface of the particles. The 24 h SSD pellets were obtained by being submerged in water for 24 h then removed from the water and rolled in a large absorbent cloth until all visible films of water were removed. Using Archimedes' principle, the density (SSD) was calculated from [19]:

$$\rho_{SSD} = \frac{m_{24\,h}}{m_{24\,h} - m_{imm}} \times 100 \quad (1)$$

The 24 h water absorption *Abs* and vacuum saturation water absorption Abs_{vac} were calculated from:

$$Abs = \frac{m_{24\,h} - m_{dry}}{m_{dry}} \times 100 \quad (2)$$

$$Abs_{vac} = \frac{m_{vac} - m_{dry}}{m_{dry}} \times 100 \quad (3)$$

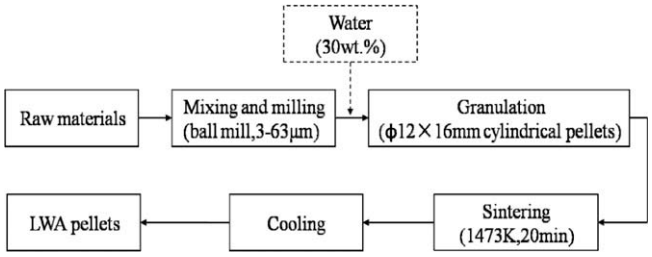


Fig. 1. Preparation of LWA pellets.

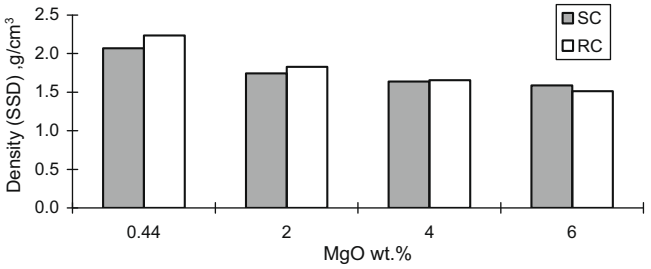
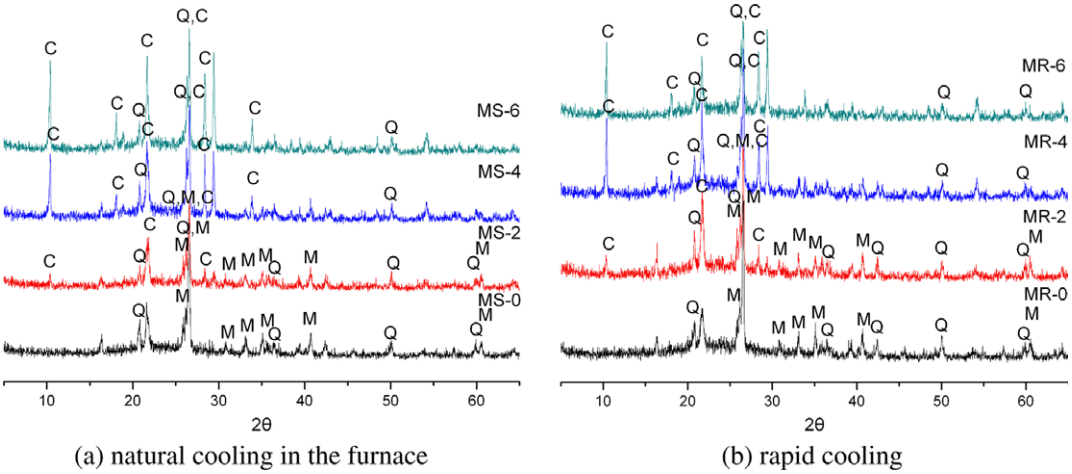


Fig. 3. Effect of MgO contents on density (SSD) of LWA pellets.



(a) natural cooling in the furnace (b) rapid cooling

Fig. 2. XRD spectra of LWA pellets with various MgO contents: C = cordierite, M = mullite, Q = quartz.

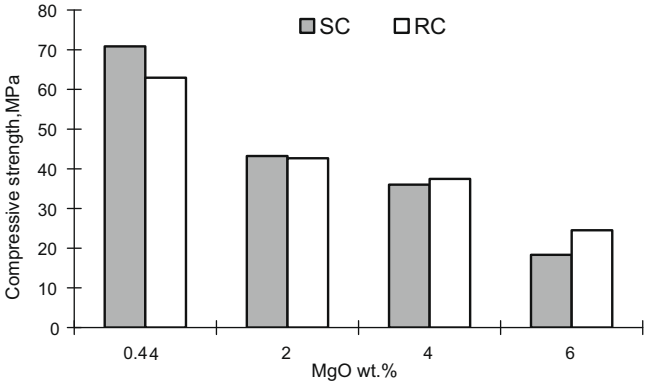


Fig. 4. Effect of MgO contents on compressive strength of LWA pellets.

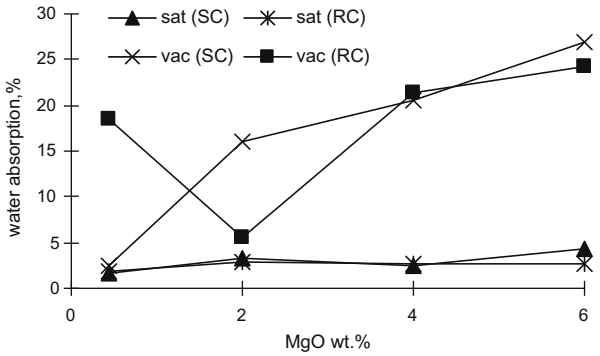


Fig. 5. Effect of MgO contents on water absorption of LWA pellets.

Mean values of compressive strength, density (SSD) and water absorption were calculated from tests completed on at least 30 pellets.

2.5. Mineralogy and microstructure analysis

The mineralogical compositions of LWA pellets were analyzed by X-ray diffraction on samples ground to less than 150 μm. Fracture surfaces of selected sintered LWA pellets were gold coated and examined using scanning electron microscopy.

3. Results and discussion

3.1. Mineralogy analysis

Phase reaction during the sintering of pellets is described in Eqs. (4)–(6). At 400–700 °C, basic magnesium carbonate is decomposed into MgO; at 400–700 °C, Kaolin is decomposed into mullite ($3Al_2O_3{\cdot}2SiO_2$) and cristobalite (SiO_2); at temperatures above 1100 °C, newly-formed mullite and that contained in fly ash reacts with MgO and SiO_2 to form cordierite.

$$3MgCO_3 \cdot Mg(OH)_2 \cdot 3H_2O \rightarrow 4MgO + 3CO_2 + 4H_2O \quad (4)$$

$$3[Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O] \rightarrow 3Al_2O_3 \cdot 2SiO_2 + 4SiO_2 + 6H_2O \quad (5)$$

$$2[3Al_2O_3 \cdot 2SiO_2] + 6MgO + 11SiO_2 \rightarrow 3[2MgO \cdot 2Al_2O_3 \cdot 5SiO_2] \quad (6)$$

The XRD spectra of LWA pellets are given in Fig. 2. This indicates that at a relatively low MgO content (at 0.44%), the primary crystalline phases are mullite and quartz. Formation of cordierite is observed at 2% MgO and the peaks of cordierite are increased at higher MgO contents, accompanied with a reduction in peaks of mullite. No detectable differences in XRD patterns are noted for the two different cooling processes.

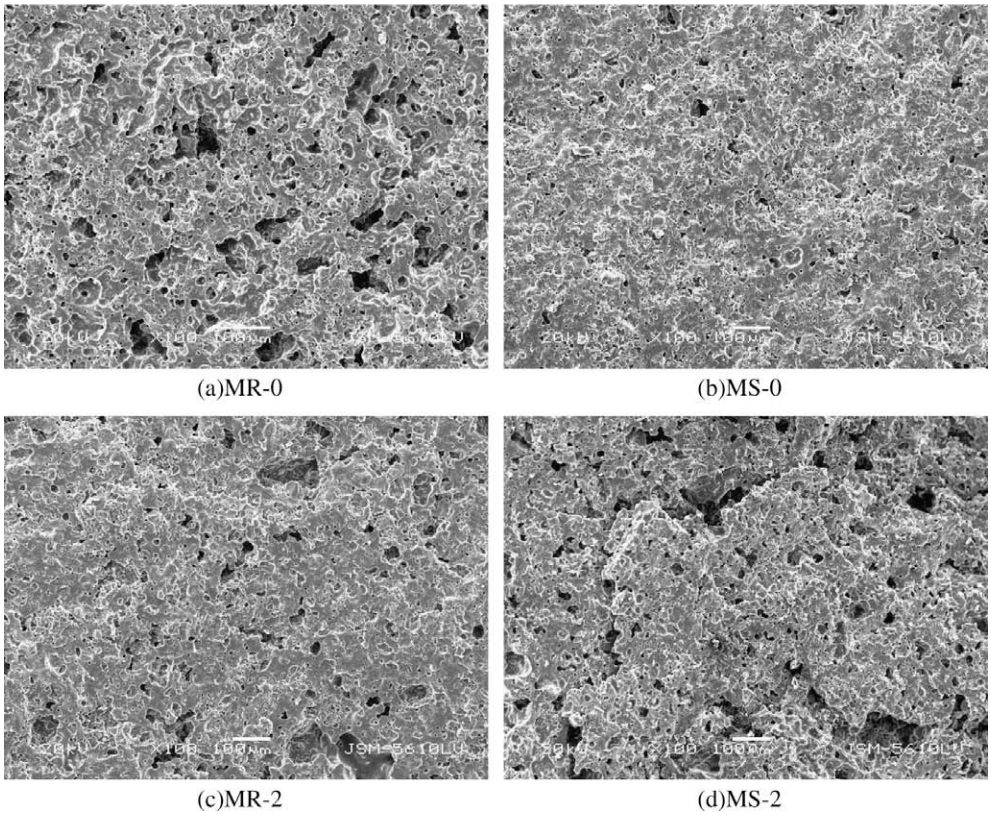


Fig. 6. SEM micrographs of LWA pellets.

3.2. Physical and mechanical properties

The effect of MgO contents on the density of LWA pellets is given in Fig. 3. More MgO produces LWA pellets with a lower density both under natural and rapid cooling. For example, the density (SSD) of pellets cooled naturally in the furnace is reduced from 2.237 g/cm^3 (at 0.44% MgO) to 1.512 g/cm^3 (at 6% MgO). This is accounted for by the decomposition of basic magnesium carbonate that generates CO_2 and H_2O. Some of the gas phase, when escaping from inside, is encapsulated by the liquid phase, resulting in the decrease in density.

It is expected that the compressive strength of LWA pellets will decrease with higher MgO contents due to the concomitant higher porosity, which is evidenced in Fig. 4. At the same time, another important reason partly responsible for the trend in Fig. 4 is that higher MgO contents result in a reduction in the content of mullite (as shown in Fig. 2), which is considered the main source of strength in LWA pellets [11–13].

It is equally worth noting in Fig. 4 that there exists a significant difference in the downward pattern of compressive strength for the two kinds of cooling processes. The compressive strength of LWA pellets decreases more steadily when they are rapidly cooled. The compressive strength of MR-0 is lower by 11.14% than that of MS-0. This level is reduced to only 1.32% when 2% MgO is present. Moreover, the compressive strengths of MR-4 and MR-6 are even higher than those of MS-4 and MS-6, respectively. It has been indicated in Fig. 2 that cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) is formed at 2% MgO, which improves the thermal-shock resistance of LWA pellets and inhibits the occurrence of cracks. Meanwhile, compressive stress resulting from rapid cooling also contributes to the increase in compressive strength of pellets.

The 24 h water absorption *Abs* and vacuum saturation water absorption Abs_{vac} of LWA pellets are given in Fig. 5. *Abs* is below 5% and does not vary distinctly under the two cooling processes. Abs_{vac} of MR-0 is 18.42% and when 2% MgO is present, Abs_{vac} of MR-2 is decreased dramatically to 5.54%. This is also accounted for by the formation of cordierite that prevents the generation of cracks and also the interconnection of pores within the pellets.

3.3. Microstructure analysis

The SEM micrographs of the inner structure of LWA pellets sintered with different MgO contents are shown in Fig. 6. When additional MgO is absent, compared to rapid cooling, natural cooling produces pellets with finer and more evenly-distributed pores, most of which are isolated. When 2% MgO is present, less connected pores are observed in pellets under rapid cooling than natural cooling. This may be because isolated pores are more likely to survive in the glassy phase.

4. Conclusions

(1) Cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) is formed at 2% MgO, and its content increases with more MgO present, accompanied by a reduction in mullite.

(2) With an increase in the content of basic magnesium carbonate in the mixture, more CO_2 is generated, resulting in a higher porosity and thus a lower density and compressive strength of LWA pellets. Compared to natural cooling, rapid cooling produces LWA pellets with more stable compressive strength and water absorption.

(3) The formation of cordierite can prevent the generation of cracks within LWA pellets, improving their strength and reducing water absorption under rapid cooling conditions for the 2% MgO system.

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