

## LOWERED TEMPERATURE RESOURCE RECYCLING OF PAPER SLUDGE USING A CO-MELTING TECHNOLOGY

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Paper sludge is a major waste by-product of the paper industry. Its disposal creates serious problems, as approximately 30% of treated sludge is not flammable. In this study, artificial lightweight aggregates (ALWAs) were synthesized from paper sludge by co-sintering with  $H_3BO_3$ .  $H_3BO_3$  acts as a flux to lower the sintering temperature below 900 °C, with co-melting occurring during the procedure. The decomposition gas is sealed within the ALWA during the glassy phase to form a porous structure. Water absorption, apparent porosity, bulk density, compressive strength, and weight loss after rinsing with  $Na_2SO_4$  were tested to understand the physical properties of the manufactured ALWAs. The optimal method suggested is co-sintering with 18%  $H_3BO_3$  flux at 890 °C for 30 min. The tested properties mentioned above gave the following results: 4.64 %, 2.77 %, 0.6 g/cm<sup>3</sup>, 13.2 MPa, and < 0.1 %, respectively. The ALWAs produced in this study have been compared to commercially available lightweight aggregates – Lytag and Arlita – with the examined ALWAs possessing better qualities than Lytag. Water absorption and compressive strength of ALWAs in this study met government requirements of pre-stressed concrete necessary for civil works, and could make useful building material.

*Keywords:* Paper sludge; Lightweight aggregate;  $H_3BO_3$ ; Co-melting

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### INTRODUCTION

Paper and pulp manufacturing involves wood preparation, pulp manufacturing, pulp bleaching, and paper manufacturing (Ochoa de Alda 2008). Wastewater coming from this process is treated by neutralization, coagulation, and sedimentation. As a result of wastewater treatment, paper sludge is the major residue, and it contains water, fiber, organic compounds, inorganic salt, and mineral fillers (Maschio *et al.* 2009). In Taiwan, the paper industry produced a total of 297,767 tons of waste sludge in 2009, and disposal of the sludge is a serious problem for the pulp and paper industries. Landfill and incineration are the primary methods of disposal. Owing to landfill space being inadequate in Taiwan, the former has become less feasible. Meanwhile, whilst incineration can reduce the volume of sludge, the non-flammable component makes up 30% of sludge (Liaw *et al.* 1998) and requires further treatment.

Paper-sludge resource recycling and associated technologies are well developed, and the sludge has many purposes; for example, as an addition to top soils in agriculture, a metal absorbent, landfill cover, and building material. In agriculture, Phillips *et al.*

(1997) carried out a 3-year experimental study on the use of paper-mill sludge on agricultural land and found that the top-soils assessed by their percentage of organic carbon were significantly improved as a result of mill-sludge applications over 3 years. Hackett *et al.* (1999) combined power boiler fly ash and wastewater paper-mill sludge composted in windrows. The final compost had a pH of 8.5, contained high concentrations of specific nutrients, and an average C:N ratio of 43:1. The dioxin concentration in the final soil/compost mixture was 3 pg/g TEQ, and the soil/compost mixture could be classified as agricultural soil. As an absorbent material for metals, Calace *et al.* (2003) employed chromatographic columns packed with paper mill sludge for metal ion removal from water. The results showed that cadmium, copper, lead, and silver could be removed from acid solutions with 100% of Cd and Cu being recovered by HCl 1.0 M; 65 % of Pb recovered by HCl 0.1 M; and 75 % of silver recovered by HNO<sub>3</sub> 0.1 M. For landfills, Moo-Young *et al.* (1997) used paper mill sludge as an impermeable landfill cover. Permeability tests showed that their samples met regulatory requirements for the permeability of landfill covers, and long-term permeability estimated from leachate-generation rates indicated that paper sludge provided an acceptable hydraulic barrier. As a recycled building material, Frías *et al.* (2008) calcined art-paper sludge for use as supplementary cementing materials in blended cement. The product calcined between 600 and 700 °C showing high pozzolanic activity. Sutcu and Akkurt (2009) added paper processing residues to an earthenware brick to produce pores. The results showed that paper processing residues decreased the density of the bricks down to 1.28 g/cm<sup>3</sup> and thermal conductivity also decreased more than 50 %.

According to Riley (1951), two conditions are necessary for the bloating of clays: (1) the clay material when heated must produce a glassy phase with a viscosity high enough to trap escaping gas; and (2) some substance must be present that liberates a gas at the temperature at which the glassy phase forms. These two principles are also essential to artificial lightweight aggregate (ALWA) manufacturing. Typically, in the use of industrial waste to manufacture ALWAs, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and flux (metal oxides such as: CaO, MgO, and Fe<sub>2</sub>O<sub>3</sub>) must be within certain content ranges for effective bloating to occur during sintering. Many types of industrial waste or sludge contain the same chemical components needed for forming lightweight aggregates. Past studies have been conducted on sewage sludge (Mun 2007), sewage-sludge ash (Lin 2006), washing aggregate sludge (González-Corrochano *et al.* 2010), mining residues (Huang *et al.* 2007), incinerated ash (Chiou *et al.* 2006), reaction ash (Chen *et al.* 2010) and reservoir sediments (Tang *et al.* 2011). While there is much research on creating ALWAs from many kinds of industrial waste, there has been relatively little on paper sludge. Paper sludge can be sintered with glass cullet or clay to produce ceramics (Asquini *et al.* 2008 and Furlani *et al.* 2011), but the mixed samples shrunk during the sintering process and the density values of the end products were more than 1.8 g/cm<sup>3</sup>. Liaw *et al.* (1998) mixed paper sludge with cement additives and the mixtures were granulated to form lightweight aggregates without sintering. The sample-density values were lower than 1 g/cm<sup>3</sup>, but water absorption was higher than 40 % in all cases. High water-absorption negatively affects the strength of lightweight concrete. For example, if the lightweight aggregates are mixed with cement to form concrete, micro-cracking occurs because the aggregates and cement have different rates of water absorption (Lo *et al.* 1999).

Co-melting technology is based on a eutectic system forming during the ceramic process. A eutectic system is one where a mixture of chemical compounds or elements has a lower melting temperature than that of either pure compound (Bi *et al.* 2003). On a phase diagram, the eutectic point represents the lowest melting point of a specific chemical composition. In other words, the melting point of a mixture can be lowered and sintered at the lower temperature by adjusting the composition of the mixture. According to past research on ALWA synthesis (Chiou *et al.* 2006; Lin 2006; Huang *et al.* 2007; Mun 2007; Wang *et al.* 2009; Chen *et al.* 2010; Kockal and Ozturan 2011), sintering temperatures for ALWAs of more than 1000 °C are required; however, high sintering temperatures are energy inefficient. In order to lower the sintering temperature, H<sub>3</sub>BO<sub>3</sub> can be added as a fluxing agent so that the viscous glassy phase can be formed at lower temperatures. H<sub>3</sub>BO<sub>3</sub> leads to dehydration in the heating process (McCulloch 1937). The dehydration of boric acid at associated temperatures is shown in Eqs.1 to 2 (Kocakuşak *et al.* 1996).



B<sub>2</sub>O<sub>3</sub> is a common flux in ceramic sintering (Soykan *et al.* 2007). For this present study, using B<sub>2</sub>O<sub>3</sub> allows sintering to occur at 890 °C.

Cheeseman and Virdi (2005) state that individual lightweight aggregate pellets should ideally have: 1) a strong but low density, porous, sintered ceramic core; 2) a dense continuous surface to avoid water absorption; and 3) a near-spheroid shape to improve fresh concrete properties. Additionally, high quality ALWAs should have low water absorption, apparent porosity, bulk density, and high compressive strength. On the other hand, according to the Chinese National Standards (CNS) of Taiwan Code 3691 and 1240, ALWAs must meet the following standards for loss on ignition and weight loss after rinsing with Na<sub>2</sub>SO<sub>4</sub> solution: less than 5 % and 12 %, respectively.

In this study, optimal sintering conditions necessary for meeting the above standards were determined for the production of ALWAs from paper sludge. Sintering temperature was reduced by the addition of B<sub>2</sub>O<sub>3</sub>. This fluxing agent allows sintering to occur at 890 °C. At this temperature gas is produced in the sintering process and trapped by the viscous glassy phase. A porous structure is formed within the sintered blocks, giving them low bulk density. The study examined all the relevant physical characteristics of the aggregates such as water absorption, apparent porosity, bulk density, weight loss after rinsing with Na<sub>2</sub>SO<sub>4</sub>, and compressive strength.

## EXPERIMENTS

### Characteristics of Paper Sludge

The paper sludge utilized in this study was from a paper mill in eastern Taiwan. To understand the sludge's water content and characteristics, a sludge sample was dried

at 110 °C and analyzed using atomic absorption spectrometry (AAS), X-Ray diffractometry (XRD), and ignition loss.

In the analyses mentioned above, 0.2 gram of dried sludge and ash were mixed respectively with 16 mL of aqua regia and 8 mL of hydrofluoric acid and heated at 180 °C for three hours. The filtrate was diluted and analyzed using AAS. An X-ray diffractometer (XRD, Rigaku) was used to examine and scan the crystalline phases of paper sludge from 10° to 80° (2 $\theta$ ) at a scan rate of 4 degrees per minute with voltage and current at 30 KV and 50 mA, respectively. For ignition-loss testing, five grams ( $W_1$ ) of paper sludge were heated at 800 °C for three hours, respectively. The weight ( $W_2$ ) of the residue was recorded while the sample was cooled. The results of ignition loss were calculated according to Eq. 3.

$$\text{Loss on ignition (\%)} = \frac{W_1 - W_2}{W_1} \times 100\% \quad (3)$$

### Experimental Procedures in the Formation of ALWAs

Extra SiO<sub>2</sub> powder was added to the wet sludge to control the ratio of SiO<sub>2</sub> versus wet sludge at 0.12. The wet sludge was dried and mixed with H<sub>3</sub>BO<sub>3</sub>. The mixture was rolled on a ball mill for 4 hours. The homogeneous mixture was shaped in a mold at 3.5 MPa for 1.5 min; then the shaped sample (2 cm in diameter) was sintered at 850 to 890 °C for 0 to 45 min. Figure 1 gives the procedural flow-diagram.

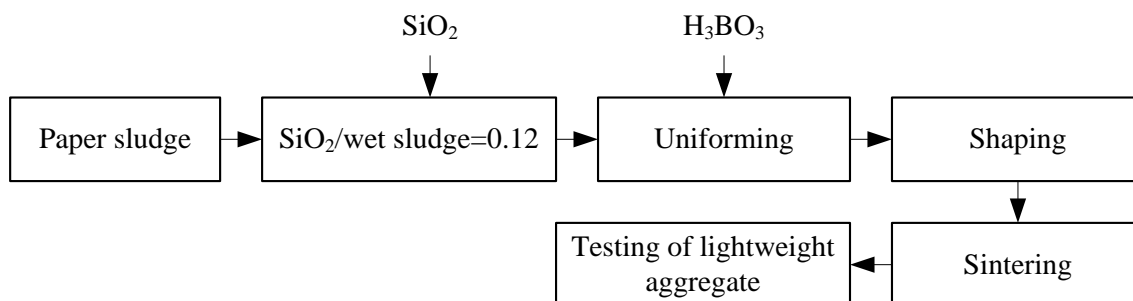


Fig. 1. Flow chart of the experiment

### Physical Tests of ALWAs

#### Water absorption

This testing was done according to CNS code 488. The ALWA was weighed ( $W_3$ ), and then soaked in water for 24 hours. After soaking, the surface of the ALWA was dried with a clean cloth, and its weight ( $W_4$ ) recorded. The water adsorption data comes from Eq. 4.

$$\text{Water adsorption (\%)} = \frac{W_4 - W_3}{W_3} \times 100\% \quad (4)$$

*Bulk density*

The ALWA was weighed in water, and the weight ( $W_5$ ) read. The volume was calculated with Eqs. 5 and 6

$$\text{Volume of sample } (V, \text{ g / cm}^3) = \frac{W_4 - W_5}{D_w} \quad (5)$$

where  $D_w$  is the density of water, and

$$\text{Bulk density } (\text{g / cm}^3) = \frac{W_4}{V} \quad (6)$$

*Apparent porosity testing*

This testing was conducted according to CNS code 619 of Taiwan. The value of apparent porosity of ALWA was calculated from Eq. 7:

$$\text{Apparent porosity } (\%) = \frac{W_4 - W_3}{W_4 - W_5} \times 100\% \quad (7)$$

*Compressive strength test*

ALWA of 2-cm diameter was tested by a single axis compression instrument, and the value calculated from Eq. 8,

$$\text{Compressive strength } (\text{MPa}) = \frac{F}{A} \quad (8)$$

where  $F$  is the compressive force and  $A$  is the aggregate cross-sectional area.

*Weight loss after rinsing with  $\text{Na}_2\text{SO}_4$  solution*

The weight loss after rinsing with  $\text{Na}_2\text{SO}_4$  solution testing procedures were in accordance with the CNS code 3618, with the results allowing us to assess material loss from the ALWA. The aggregate sample was weighed ( $W_3$ ), and put into a saturated solution of sodium sulfate for 16 hours. Then the sample was dried in an oven until the sample weight was constant. The procedure was repeated 5 times, and the final sample was cleaned with distilled water at  $43 \pm 6$  °C. The filtrate of the final sample was examined with a solution of barium chloride to ensure that sodium sulfate was removed completely. The sample was again dried to constant weight and the weight recorded ( $W_6$ ). The value of weight loss was calculated using Eq.9,

$$\text{Weight loss after rinsing with } \text{Na}_2\text{SO}_4 \text{ solution } (\%) = \frac{W_1 - W_6}{W_1} \times 100\% \quad (9)$$

*Loss on ignition of ALWAs*

In accordance with CNS 1078, ALWAs were ground until the particle size was less than 1 mm. 1 g (*W7*) of ALWAs powder was heated at 950 °C for three hours. The weight (*W8*) of the residue was recorded while the sample cooled. The result of loss on ignition was calculated according to Eq.10:

$$\text{Loss on ignition (\%)} = \frac{W7 - W8}{W7} \times 100\% \quad (10)$$

*Scanning electron microscope (SEM) analysis*

A Jeol JSM6360 high resolution SEM was used to observe the surface and cross section morphology of the samples. The size of the specimen was about 0.2×0.2 cm<sup>2</sup>, and it was sprinkled so as to allow platinum gilding by vacuum deposition.

**RESULTS AND DISCUSSION****Characteristics of Paper Sludge**

The results of analysis of the paper sludge utilized in this study are shown in Table 1. The water content was 65%, and loss on ignition of the dry sludge was 52.63%. High values of loss on ignition implied high organic content due to fiber or other organic compounds. Chemical composition is also shown in Table 1, with the major element of sludge being Ca. The calcium content might arise from the neutralizer used in wastewater treatment or papermaking filler used in the papermaking process (Ochoa de Alda 2008). The result of XRD analysis (see Fig. 2) matched the chemical analysis and showed that the major phase of sludge was (Ca,Mg)CO<sub>3</sub>.

**Production of ALWAs***Addition of SiO<sub>2</sub>*

SiO<sub>2</sub> was added to the wet sludge to control the ratio of SiO<sub>2</sub> to wet sludge at 0.12 (hereafter referred to as conditioned paper sludge). The addition of SiO<sub>2</sub> promotes aggregate bloating and the viscous glassy phase during sintering. The glassy phase is able to trap the gas within the sample. Tsai *et al.* (2006) investigated the characteristics of lightweight aggregates sintered from sewage sludge ash by modifying the proportion of

**Table 1.** Analysis of Paper Sludge

	Water content (%)	CaO	SiO <sub>2</sub>	MgO	Al <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	Loss on Ignition (%)
paper sludge	65.0	20.90	16.70	1.81	2.10	1.22	3.86	52.63

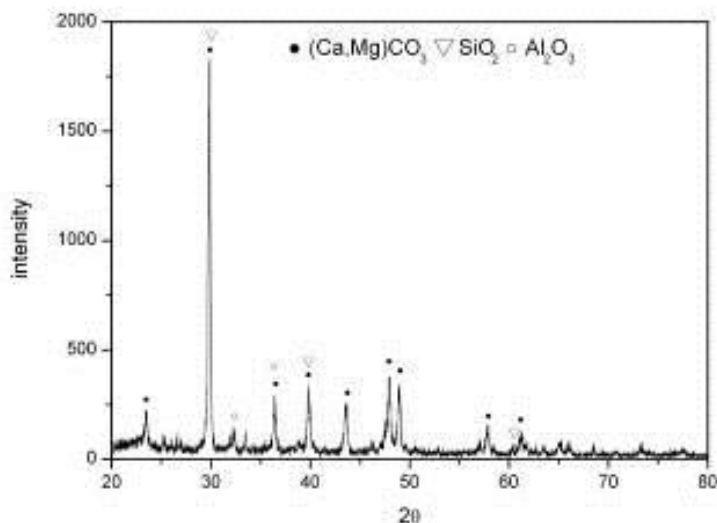


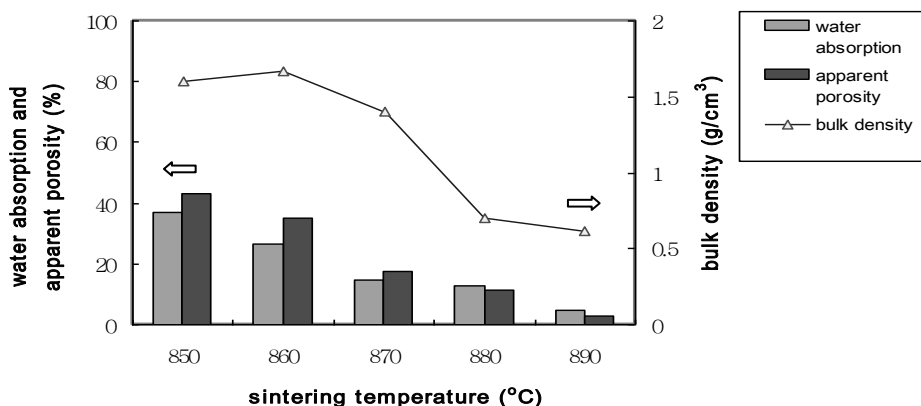
Fig. 2. XRD analysis of paper sludge

the main components ( $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-flux}$ ). According to their results, the addition of  $\text{SiO}_2$  had an obvious effect on the swell rate of the lightweight aggregate. The swell rate increased from 70 % to 110 % with the addition of  $\text{SiO}_2$  (0% to 20%) at a sintering temperature of 1080 °C. Density also dropped with increasing  $\text{SiO}_2$  dosage. The authors also state that sticky glassy materials formed at the surface to promote more gas being trapped within the aggregate. In this present study, paper sludge contained 16.7%  $\text{SiO}_2$  (see Table 1), and the increase in  $\text{SiO}_2$  promoted bloating in the sintered paper sludge.

#### Effect of sintering temperature on ALWAs

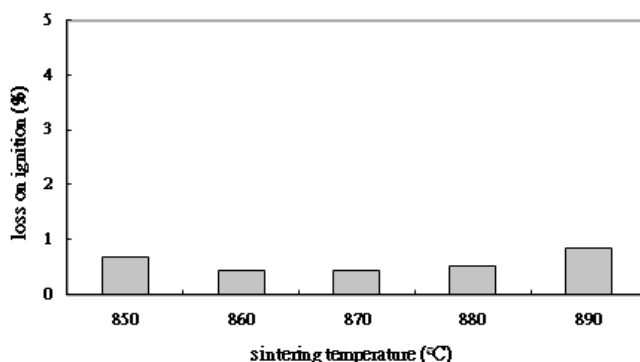
The conditioned paper sludge was mixed with 18 %  $\text{H}_3\text{BO}_3$ . The sintering temperature was set between 850 to 890 °C for 30 min. When the temperature was increased from 850 °C to 890 °C, the values of water absorption, apparent porosity and bulk density decreased from 36.7 % to 4.6 %; 43.1 % to 2.8 %; and 1.6  $\text{g/cm}^3$  to 0.6  $\text{g/cm}^3$ , respectively (see Fig. 3). In our experiments, the water absorption rate was affected by the size and quantity of pores on the aggregate's surface (Huang *et al.* 2007). Water absorption and apparent porosity decline simultaneously; *i.e.*, the volume of water that can penetrate an aggregate varies positively with the surface-connectivity of pores.

Our experimental results are similar to those of Cheeseman *et al.* (2005) in that aggregates sintered at high enough temperatures had surfaces that were sealed by the glassy phase. Additionally, the value of bulk density also decreased significantly with sintering temperatures above 870 °C. The reduction in bulk density indicates the occurrence of bloating and gases being trapped within the sample, *i.e.*, the presence of the glassy phase sealing the sample's surface. At temperatures lower than 870 °C, gases could escape through the unsealed open pores giving a bulk density of the final aggregate of more than 1  $\text{g/cm}^3$ . Variation in surface and inner pores is presented by SEM analyses and discussed below.



**Fig. 3.** Physical properties of ALWAs sintered at different temperature for 30 min ( $H_3BO_3$  / sludge=0.18)

In addition to the aggregate's physical properties, the CNS 3691 regulates that loss on ignition of lightweight aggregates must be less than 5 %. Paper sludge contains organic components such as wood fibers, pitch, and lignin by-products (Ochoa de Alda 2008). These organic compounds must be eliminated to meet the regulatory standard. The results of sintering ALWAs at different temperatures are shown in Fig. 4. The values of all ALWAs are lower than 1%, which means that most of the organic compounds in the paper sludge were burned off during the sintering process. These analyses show that a temperature greater than 850 °C is needed to burn off organic compounds, but to produce an aggregate with the appropriate physical properties a sintering temperature of 890 °C is required (see Fig. 3).



**Fig. 4.** Loss on ignition of ALWAs sintered at different temperature for 30 min ( $H_3BO_3$  / sludge=0.18)

#### *Effect of the dosage of $H_3BO_3$ in ALWAs*

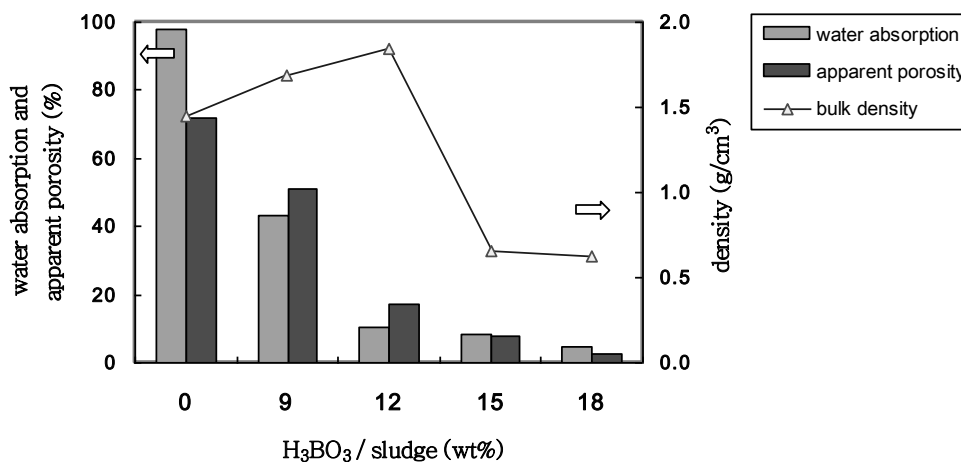
As discussed in the Introduction, the addition of flux can reduce the melting point of sludge due to the creation of a eutectic system during sintering. A eutectic system is one where the co-melting point of the mixture of chemical components (of sludge) is lower than that of individual components. When  $H_3BO_3$  is used as a flux, it dehydrates to



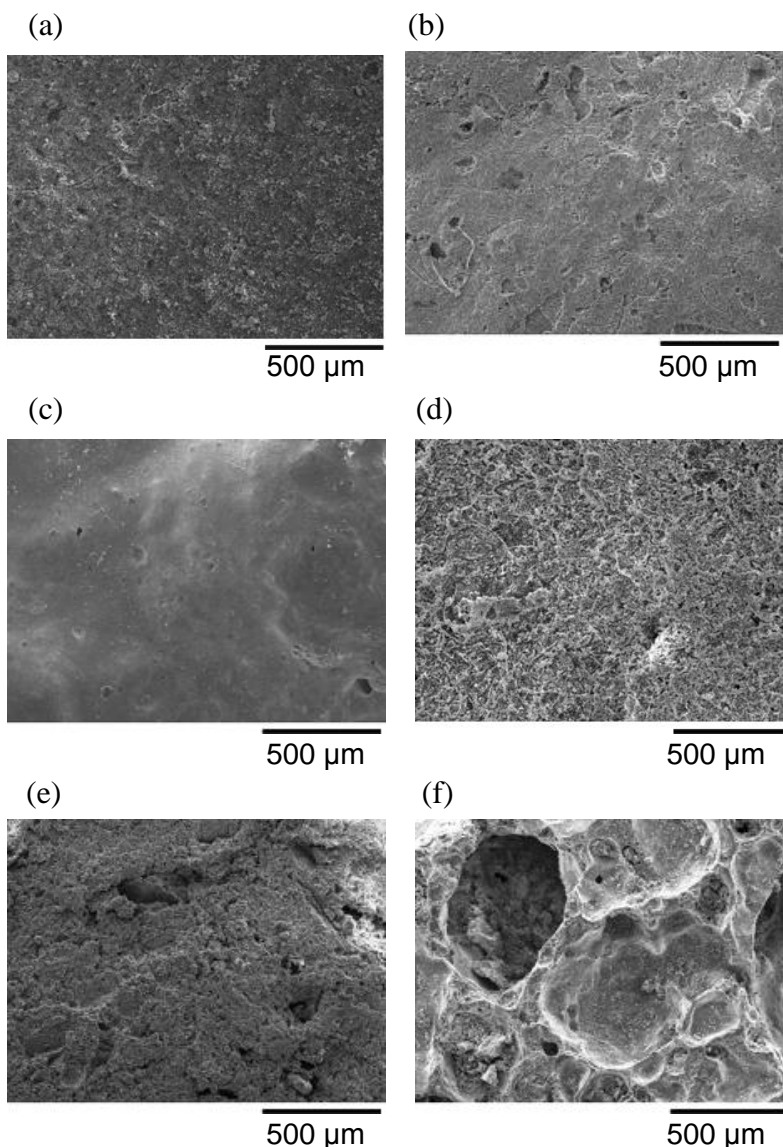
$B_2O_3$ , which is a low melting point compound. In our study, the addition of  $H_3BO_3$  lowers the co-melting point of paper sludge and promotes the formation of the viscous glassy phase. The glassy phase seals surface pores, trapping gas and bloating the sample. ALWAs with soundly formed glassy phases have low water absorption, apparent porosity, and bulk density. In this study, the values of water absorption and apparent porosity dropped from 97.8 % to 4.64 % and 71.8 % to 2.77 % when  $H_3BO_3$  dosage was increased from 0 wt% to 18 wt%. Additionally, bulk density also decreased from 1.84  $g/cm^3$  to 0.6  $g/cm^3$  when the dosage was increased from 12 wt% to 18 wt% (see Fig. 5).

At  $H_3BO_3$  dosage of less than 15 wt%, the glassy phase did not form soundly, and gas could escape through non-sealed surface pores. As a result, water absorption, apparent porosity, and bulk density of the ALWA were high. However, at higher  $H_3BO_3$  dosing the glassy phase was well formed. The surface pores were apparently sealed, and gas was trapped in the ALWA. It is the combination of sealed surface pores, but apparent inner pores that gives the ALWA its favorable characteristics of low water absorption and bulk density.

The SEM images of ALWAs produced for doses of 0 wt% to 18 wt% are shown in Fig. 6. In Figs 6(a), (b) and (c), the surfaces of aggregates produced from sludge dosed with 18 wt% of  $H_3BO_3$  were much smoother than the others; the smooth surface indicates vitrification being more complete (Huang *et al.* 2007). Figures 6(d), (e), and (f) show that large inner pores were only formed when boric acid dosage levels were high. The formation of inner pores indicates gas being trapped by the glassy phase. This greatly reduces bulk density. Mueller *et al.* (2008) studied the expansion of granules from zeolitic rocks. The ground rocks were mixed with soda ash ( $Na_2CO_3$ ) in order to lower their sintering temperature. They found that the soda content improved the formation of inner pores, and the pore size increased with increased soda dosage. This is a similar phenomenon to that which is occurring with the paper sludge. On the other hand, bulk density increased for doses between 0 wt% to 12 wt%. In this case, the glassy phase was not properly formed to trap gasses and increased flux promoted densification (Teo *et al.* 2008; Soykan 2007; Soykan *et al.* 2006).



**Fig. 5.** Physical properties of ALWAs sintered at different dosage of  $H_3BO_3$  at 890°C for 30 min

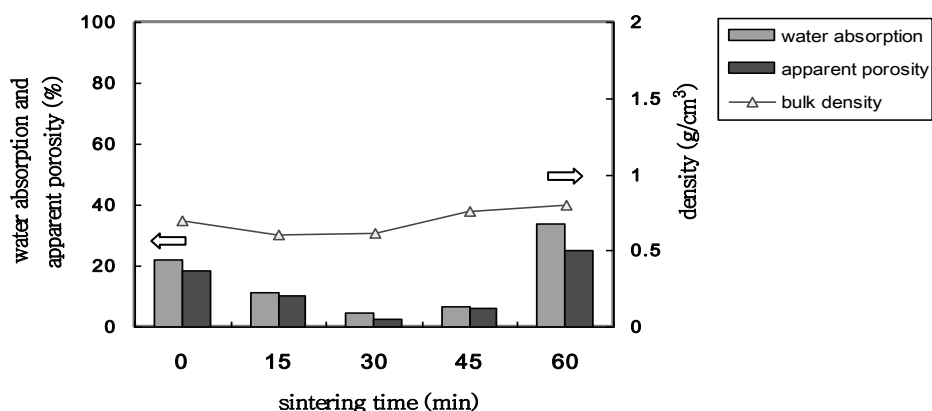


**Fig. 6.** SEM images of ALWAs sintered with different dosage of  $H_3BO_3$  at 890 °C for 30 min. (a) Surface,  $H_3BO_3$ /sludge = 0 (b) Surface,  $H_3BO_3$ /sludge = 0.09 (c) Surface,  $H_3BO_3$ /sludge = 0.18 (d) Cross-section,  $H_3BO_3$ /sludge = 0 (e) Cross-section,  $H_3BO_3$ /sludge = 0.09 (f) Cross-section,  $H_3BO_3$ /sludge = 0.18

#### *Effect of the sintering time in ALWAs*

Sintering time is an important procedural aspect of forming ALWAs. In this case sintering times from 0 to 60 min were investigated. Both 0- and 60-min sintering times were associated with high water absorption and apparent porosity for different reasons (see Fig. 7). Figures 8(a) to (e) show SEM images of ALWAs sintered for 0 to 60 min and the accompanying variations in pore size and number. Note that surface pores are apparent for the earlier and latter sintering times and less evident for the mid-range sintering times. This is because at a sintering time of 0 minutes the viscous glassy phase has not had a chance to form and surface porosity is high. On the other hand, in the case

of longer sintering times the glassy phase becomes more fluid, and the trapped gasses expand to the point where they pop through the liquid glass surface. This leads to pores at the surface of the aggregate. Hung *et al.* (2007) describes a similar phenomenon. The trend in bulk density follows a similar pattern to that of apparent porosity and water absorption. Bulk density first decreases with time as gasses are trapped by the viscous glassy phase at the surface of the aggregate but then increases as the glassy phase turns more fluid and the gasses are able to escape. The escaping gasses allow for the glassy phase to fill the inner pores with liquid glass, resulting in increased bulk density. Figure 9 shows shrinkage of the inner pores with increased sintering time. These results show that an ideal sintering time for conditioned paper sludge is 30 min (see Fig. 7).



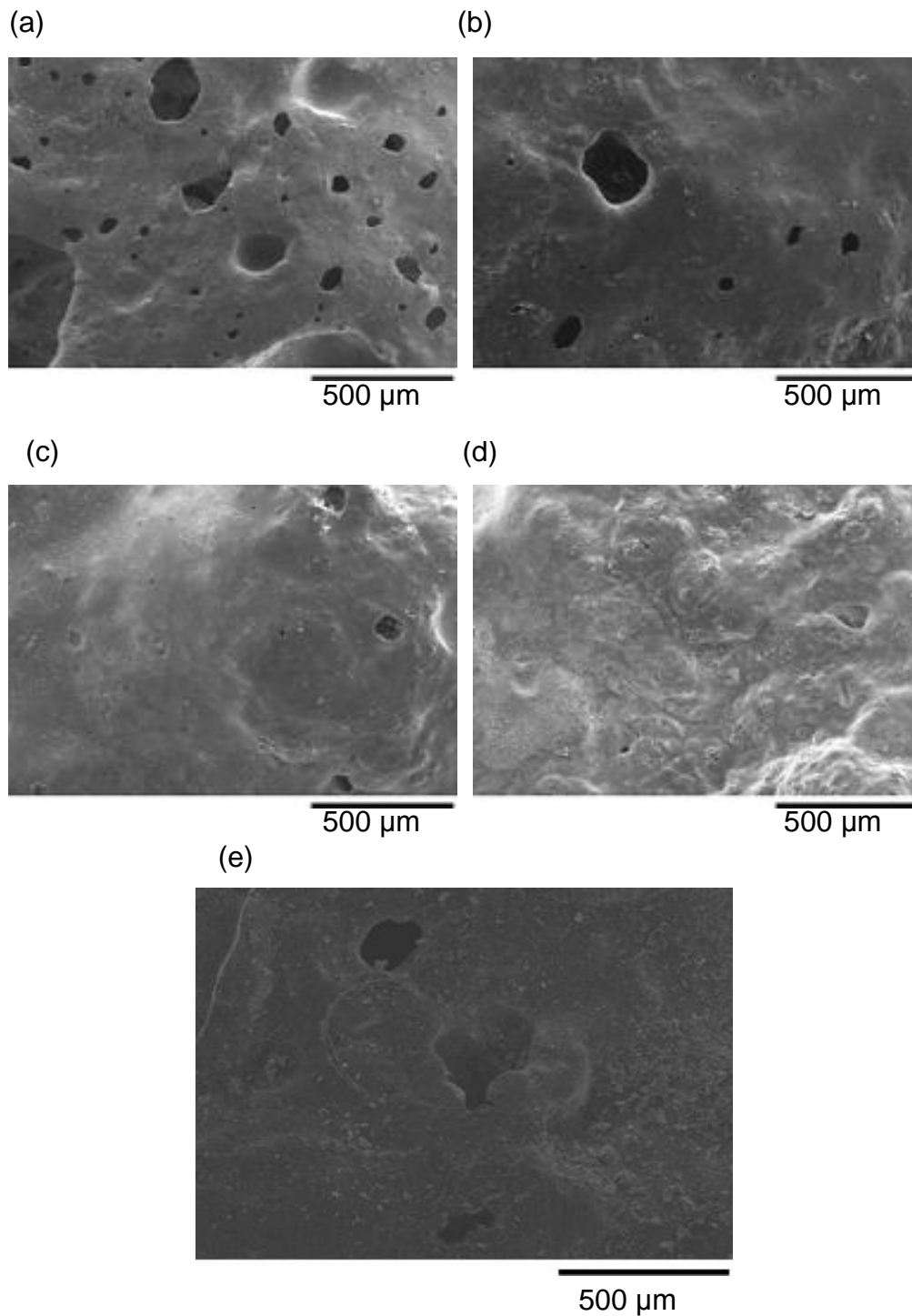
**Fig. 7.** Physical properties of ALWAs sintered at 890 °C for different times ( $H_3BO_3$ /sludge=0.18)

#### *Compressive strength and weight loss after rinsing with $Na_2SO_4$ solution*

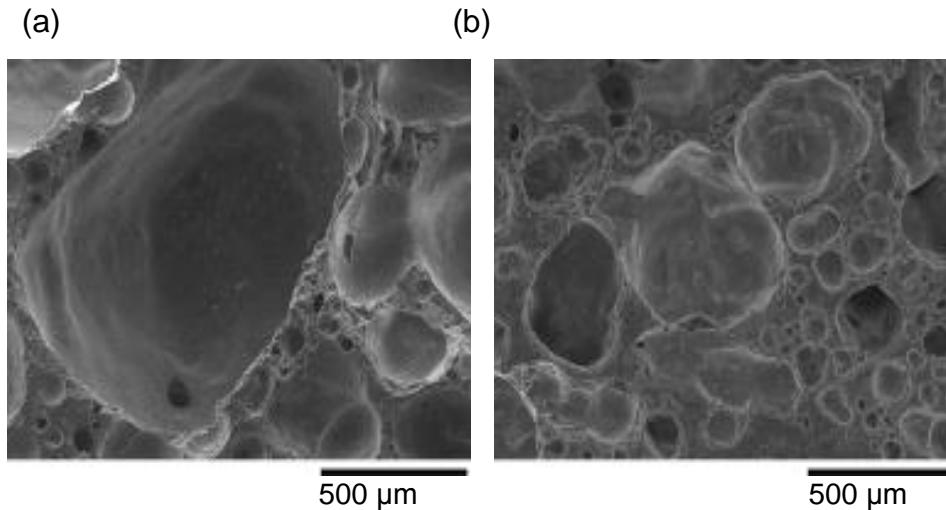
Compressive strength is another very important attribute of ALWAs if they are to be used as construction materials (Tsai *et al.* 2006). ALWAs sintered at 890 °C for 15, 30, and 45 min were tested to understand if a trend existed between sintering time, compressive strength, and weight loss after rinsing with  $Na_2SO_4$  solution. The results of these tests are shown in Table 2. The values of compressive strength ranged between 9 (MPa) and 13 (MPa). In this case, the strength of the paper-sludge ALWA was higher than Lytag, a commercially available lightweight aggregate, whose compressive strength ranges between 5 (MPa) and 9 (MPa) (Cheeseman *et al.* 2005). The results of weight loss for all samples were below 0.1 %, and they met the regulatory limit of 12%. The low value indicates that the aggregates can resist water expansion under a variety of temperatures.

**Table 2.** Compressive Strength and Weight Loss after Rinsing with  $Na_2SO_4$  Solution of ALWAs Sintered at 890 °C for Different Rimes (with 18%  $H_3BO_3$ )

Sintering time (min)	Compressive strength (MPa)	Weight loss (%)
15	9.7	<0.1
30	13.2	<0.1
45	11.6	<0.1



**Fig. 8.** Surface SEM images of ALWAs sintered at 890 °C for different times ( $H_3BO_3$ /sludge=0.18) (a) 0 min (b) 15 min (c) 30 min (d) 45 min (e) 60 min



**Fig. 9.** Cross-section SEM images of ALWAs sintered at 890 °C for (a) 5 min and (b) 45 min

#### *Comparison and application of ALWAs*

Optimal sintering parameters were chosen based on the aforementioned positive physical properties of ALWAs. These are: dosage percentage of  $H_3BO_3$  at 18% of conditioned sludge, temperature of 890 °C, and dwell time of 30 min. For these parameters the aggregate exhibited its lowest water absorption, apparent porosity, and highest compressive strength. The values of these properties are shown in Table 3. Water absorption and density of the ALWA were lower than Lytag, and compressive strength was higher. The favorable physical properties of this ALWA were due to the formation of the viscous glassy phase, which was promoted by  $H_3BO_3$ .

**Table 3.** Comparison between ALWAs in this Study and Lytag

Physical Properties	ALWA	Lytag
Water absorption (%)	4.64	10-17
Apparent porosity (%)	2.77	--
Density (g/cm <sup>3</sup> )	0.6	1.35-1.5
Compressive strength (MPa)	13.2	5-9

The characteristics of another commercial aggregate, Arlita, are shown in Table 4 (González-Corrochano *et al.* 2009). Arlita has a range of varieties based on the physical properties of each type. These are given by G3, F3, F5, and F7 in Table 4. Lightweight aggregates with higher compressive strength and lower water absorption are utilized in buildings and concrete slabs. The aggregate produced from paper sludge in this study compares favorably with Arlita F7, meaning it is suitable as a construction material.

**Table 4.** Varieties of Arlita, Features and Application (González-Corrochano, 2009)

Variety of Arlita	Water absorption (%)	Compressive strength (MPa)	Application
G3	20	0.981	Insulation, geotechnical applications, gardening and horticulture
F3	20-25	1.962	Prefabricated lightweight structures and insulation lightweight concretes
F5	15-20	4.905	Concrete slabs, building structures
F7	10-15	6.867	Prestressed concrete, civil works

#### *Cost-Benefit assessment of produced ALWA*

A cost-benefit assessment for current treatment methods and the proposed ALWA for paper sludge are shown in Table 5. The disposal cost of paper sludge is about 2500 NT dollars per ton in Taiwan. We base our comparative assessment on a lightweight aggregate sintered with 18 wt% of H<sub>3</sub>BO<sub>3</sub> at 890 °C for 30 min. All values are given per ton of paper sludge. SiO<sub>2</sub> and H<sub>3</sub>BO<sub>3</sub> usage per ton was 93.3 kg and 79.7 kg, respectively, and the ALWA yield was 395 kg per ton of sludge. The price of ALWA depends on aggregate volume, so it is necessary to know dry loose bulk density. The dry loose bulk density of paper sludge ALWA is calculated by Eq. 11.

$$\text{Dry loose bulk density} \left( \frac{\text{kg}}{\text{m}^3} \right) = A \times \rho \times 1000 \quad (11)$$

The constant *A* is the ratio of dry loose bulk density versus particle density. According to other researchers (Tay *et al.* 2003; Chen *et al.*; Tang *et al.* 2011), *A* is between 0.43 to 0.62; it was set at 0.6 as Chen *et al.* (2010), because our paper sludge aggregate's particle density was similar to their product. The parameter  $\rho$  (g/cm<sup>3</sup>) is the bulk density of ALWA, and we set this value at 0.6 g/cm<sup>3</sup>. As a result, the dry loose bulk density was 360 kg/m<sup>3</sup>. Therefore, in our experiments one ton of paper sludge yields 1.1m<sup>3</sup> (*i.e.* 395 kg) of ALWA. The price of ALWA is 2500 to 4000 NT/m<sup>3</sup>, and an average price of 3250 NT/m<sup>3</sup> is used. Energy consumption is given as kilowatt-hour (kWh), and energy cost is calculated based on different energy sources such as electricity, heavy oil, and coal.

The current cost of disposal is \$2500NT. This cost could be saved if ALWA were manufactured from paper sludge. Overall, cost-benefit assessment is most influenced by the price of energy. The price of electricity and heavy oil are 4.4 and 3.7 times that of coal. A net benefit of \$2061NT could be achieved if the cost of current disposal is considered a saving and coal is used as an energy source for sintering.

**Table 5.** Cost-Benefit Assessment of Current Treatment and Co-Melting Technology <sup>(1)</sup>

Disposal method	Assessment items	Cost	Benefit
Current treatment	Commissioned disposal (A)	2500	-
ALWA product	Agents cost <sup>(2)</sup>	SiO <sub>2</sub> (B)	56
		H <sub>3</sub> BO <sub>3</sub> (C)	2359
	Energy cost <sup>(3)</sup>	Electricity (D1)	6811
		Heavy oil (D2)	5858
		Coal (D3)	1563
	Subtotal cost	(B)+(C)+(D1)	9226
		(B)+(C)+(D2)	8309
	(B)+(C)+(D3)	4014	-
	ALWA benefit (G) <sup>(4)</sup>		3575
Net benefit:		(A)+(G)-(B)-(C)-(D1)	-3151
		(A)+(G)-(B)-(C)-(D2)	-2234
		(A)+(G)-(B)-(C)-(D3)	2061

<sup>(1)</sup> Unit: NT\$ per ton paper sludge; Exchange rate: US\$ 1.00 was about NT\$ 29.5

<sup>(2)</sup> The prices of SiO<sub>2</sub> and H<sub>3</sub>BO<sub>3</sub> were NT\$ 0.6 and 29.6 per kilogram, and the amount of these two agents were 93.3 kg and 79.7 kg per ton paper sludge, respectively.

<sup>(3)</sup> The prices of electricity, heavy oil and coal were NT\$ 2.365, 2.03 and 0.543 per kilowatt-hour and energy consumption was 2880 kWh per ton sludge.

<sup>(4)</sup> One ton of paper sludge could produce 395 kg ALWA *i.e.*, 1.1 m<sup>3</sup> of dry loose aggregate (Eq. 12). The price of ALWA is NT\$3250 per cubic meter.

## CONCLUSIONS

1. Paper sludge contains inorganic filler; it is suitable for use as a raw material in producing sintered aggregates.
2. The formation of the viscous glassy phase is improved by H<sub>3</sub>BO<sub>3</sub> addition. The ideal level of flux to conditioned sludge is 18 wt% of H<sub>3</sub>BO<sub>3</sub>. This level of flux allows for sintering to occur at a lowered temperature of 890 °C, with the viscous glassy phase forming at the surface of the aggregate to trap gasses. This situation contributes to the favorable aggregate characteristics of decreased water absorption, apparent surface porosity, and bulk density.
3. The optimal sintering conditions were 18 wt% of H<sub>3</sub>BO<sub>3</sub> at 890 °C for 30 min, and the physical properties of ALWA produced were 4.64 % water absorption, 2.77 % apparent porosity, 0.6 g/cm<sup>3</sup> bulk density, and 13.2-MPa compressive strength. The loss on ignition and weight loss after rinsing with Na<sub>2</sub>SO<sub>4</sub> solution were less than 5 % and 12 %, which met the regulatory standards of Taiwan.
4. Cost-benefit assessment of the manufacture of a paper-sludge ALWA showed that a net benefit of \$2061NT could be achieved if the cost of current disposal is considered a saving and coal is used as an energy source for sintering.

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