

Consolidation of Incineration Fly Ash by Solvothermal Reaction

Kaoru Masuda and Shigehisa Endoh

Materials Utilization Group, Research Institute for Green Technology
National Institute of Advanced Industrial and Technology (AIST)
16-1 Onogawa, Tsukuba-shi, Ibaraki, 305-8569, Japan

The generation of fly ash tends to increase yearly so that this is currently considered a big environmental concern, which requires appropriate treatment approaches. In this research the consolidation of incineration fly ash by the hot-press solvothermal reaction was investigated to provide an alternative process for the treatment and utilization of this waste material. Results showed that at reaction conditions of 52 K temperature, 20 MPa pressure and 60 minutes treatment time, the resulting consolidate exhibited a compressive strength of 37-40MPa, a tensile strength of 6.5-7.0 MPa and a Rockwell hardness of 20-23 RH15W.

These properties are comparable to the compressive strength of Portland cement, which ranges from 30-40 MPa as well as with the tensile strengths of mortar, granite, artificial lightweight aggregate and solidified high concrete whose values are 2-2.5 MPa, 5-9 MPa, 5-10 MPa and 3-5 MPa, respectively.

Furthermore, by mixing fly ash with glass at 50% ratio and then subjecting to similar treatment conditions, a consolidate with even higher tensile strength of 12.5-13.3 MPa and hardness of 77-80 RH15W may be achieved.

Keyword: incineration fly ash, heavy metal, solvothermal hot-press, solvothermal reaction

1. Introduction

The amount of fly ash generated from the incineration of municipal waste tends to increase yearly resulting to serious disposal and treatment problems [1]. As the incineration fly ash contains various toxic heavy metals, preliminary removal of these elements is indispensable when reclamation and recycling concerns are taken into consideration [2].

At present, cement solidification is employed as one of the effective recycling method for incineration fly ashes. Unfortunately, this process has problems on the possibility of secondary pollution by the infiltration of heavy metals due to their chronological changes [3].

Although the adsorption using chelating agent, thermal treatment and plasma treatment have been studied for the treatment of the incineration fly ash, some cost-related problems remain. For appropriate treatment and utilization of heavy metal-containing incineration fly ash, safe and profitable technologies are needed.

In this research the consolidation of incineration fly ash by the hot-press solvothermal reaction was investigated to provide an alternative process for the treatment and utilization of this waste material.

Initial investigations on the optimization of hydrothermal

hot-press method for effective use of incineration fly ash as a structural or construction material are also conducted

2. Features of Solvothermal Reaction

One of the features of a high temperature and high-pressure water solution treatment is said to be the possible acceleration of ion reaction and hydrolysis. When heated to higher than 273K in an airtight vessel, ion reaction is induced and promoted [4]. The ion product value ($[H][OH]$), which is one of the factors dominating the chemical nature of water, is 10-14 at 298 K [5]. This value changes in accordance with the rise of temperature under the saturated vapor pressure.

The ion product is maximum at around 573 K, which are several thousand times higher than at room temperature. It decreases at temperatures higher than 573 K since the condition of water then changes into gaseous state.

Therefore, the ionic reaction at around 573 K is considered to be ideal for the dissolution of heavy metals existing in the incineration fly ash [5]

3. Experimental

A conceptual flow sheet of the incineration fly ash treatment is shown in Fig. 1.

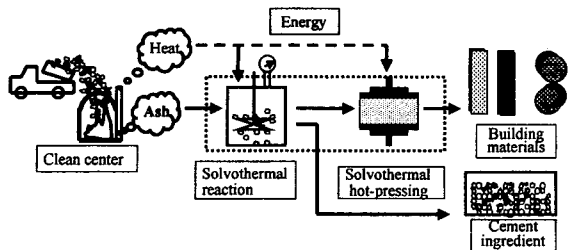


Fig.1 Conceptual flow sheet of the incineration fly ash treatment

Analysis showed that the heavy metals such as Zn, Cd, Pb, Cu and Hg were highly concentrated in the fly ash (Table 1).

Table 1 Main of composition in incineration fly ash

Composition	Si	Ca	Fe	Al	Na	K	Mg	Mn
Quantity	13.20	17.22	0.67	4.74	3.37	3.53	1.32	0.02

(unit:w%)

This is probably due to their lower melting point and higher vapor pressure. Analysis of the fly ash have also revealed the presence of Na, K, Ca cations, SO₄, and Cl anions as well as traces of the more toxic metals Hg, Cd and Cr. By the thermal quantity reduction method, the amount of the unburnt portion in the fly ash was determined to be about 6 % in weight.

Figure 2 shows the schematic diagram of the set-up employed during the solvothermal-hot press treatment of incineration fly ash.

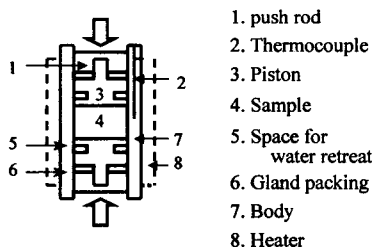


Fig.2 Autoclave for solvothermal Hot-pressing

The equipment is made up of stainless steel cylinder of 300 Mm length and 27 mm inner diameter. It is equipped with Teflon packing with movable pistons at the top and bottom of the cylinder. Through the actions of the pressing rod on the pistons, the pressure inside the cylinder is increased, which leads to subsequent solidification of the samples.

The equipment is also equipped with a heater as well as a thermocouple for varying and monitoring the reaction temperatures. During the experiments, 10 gm each of pure incineration Fly ash as well as mixtures of incineration fly ash and glass (at10, 25 and 50%) was combined with 10%

Distilled water.

The samples were transferred into the autoclave and Treatment runs at temperatures of 473 and 523 K and reaction times of 10, 30, 60 and 120 minutes while maintaining a constant pressure of 20 MPa. Heat addition was set at a high rate, which results to a temperature rise of approximately 278K/min.

The consolidate products were removed from the autoclave and subjected to different physical and chemical analysis.

The tensile strength of the consolidate samples were obtained using the following equation:

$$\sigma = 1/2. P_{max}. d. t,$$

Where P_{max} = maximum load, d = diameter and t = thickness.

A material testing machine (TOYO-BOLDIN Co., Ltd., model LTM-10) was used to measure the compressive strength and tensile strength of the sample in accordance with the Japan Industrial Standard.

The hardness of the samples was measured by a Rockwell machine (modeleSHT-32,AkashiCo.,Ltd).

SEM analysis was also conducted (model JSM-5410, Nihon Denshi Co., Ltd.) was also made to evaluate the physical and chemical changes that occurred in the sample at different reaction conditions.

4. Results and Discussions

The values of the specific gravity of the consolidated samples were almost uniform at 2.1-2.2 x 10³ kg/m³.

Figure 3 shows the relation between the applied temperature and the compression strength of consolidated sample at a reaction time of 60 minutes and applied pressure of 20 MPa. Higher reaction temperature results to an enhanced compression strength of the sample.

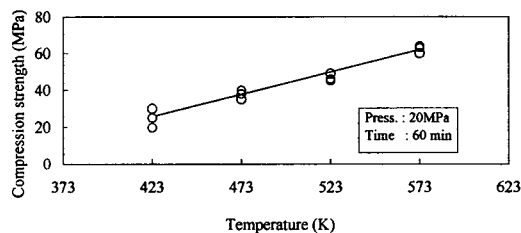


Fig.3 Relation between reaction temperature and compression strength of consolidated sample

At a reaction temperature of 473 K, the compressive strength was approximately 38 MPa, which is almost comparable to that of normal concrete [6]. By increasing the reaction temperature to 573 K, the compression strength increased to as high as 60 MPa. Results from other runs showed that Compression strength of 28 MPa and 75 MPa may be

Compression strength of 28 MPa and 75 MPa may be Reaction times of 20 minutes and 120 minutes, respectively.

For the effective use of incineration fly ash as a structural or construction material.

Figure 4 shows the relation between reaction time and physical properties of the consolidated sample at an applied s

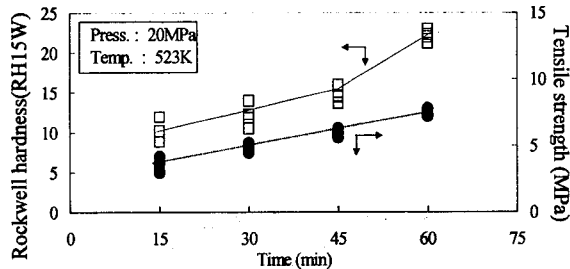


Fig.4 Relation between rockwell hardness, tensile strength and reaction time of consolidated sample

Solidification pressure of 20 MPa and reaction temperature of 523 K. At 15 minutes solidification time and 523 K reaction temperature, the tensile strength of the consolidated sample was approximately 3.7-4.2 MPa.

This is approximately 3-4 times the value at 273 K, which is about 0.7-1.1 MPa. By increasing the reaction time to 60 minutes, the tensile strength increased to approximately lightweight aggregate and solidified high concrete waste are 5-10 MPa and 3-5 MPa, respectively [7]

From Figure 4 also, the hardness of the consolidated sample at 15 minutes treatment time was found to increase from 1.5-4 RH15W at 273 K to 7-12 RH15W at 523 K reaction temperature. At 60 minutes, the hardness increased to 22 RH15W.

Since the hardness value was measured at different points on the surface of the sample, the wider range of value at 15 minutes suggests the insufficiency of this reaction time. Approximately 60 minutes is necessary for reaction to Proceed to completion in order to produce a material of Uniform hardness. Furthermore, the effect of mixing fly

Ash with glass powder on the compression strength, tensile strength and Rockwell hardness of the consolidated sample was also studied. At 50 % mixing ratio, the Rockwell hardness was increased from 23 RH15W at zero mixing ratio to 80 RH15W (Figure 5).

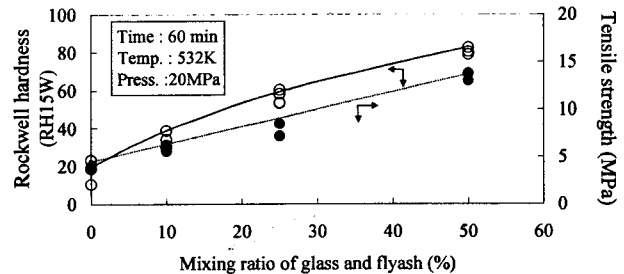


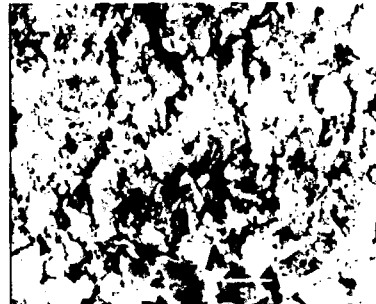
Fig.5 Relation between rockwell hardness, tensile strength and mixing ratio of glass and flyash of consolidated sample

For reference, the hardness of plastic materials for the manufacture of car bearings (placental) is about 82 RH15W. With regards to the tensile strength, glass mixing at 50 % ratio increases hardness to 13.2 MPa from its value of 6.5-7 MPa at zero glass mixing. The values of these properties are comparable to those of artificial aggregates (10-15 MPa) [7].

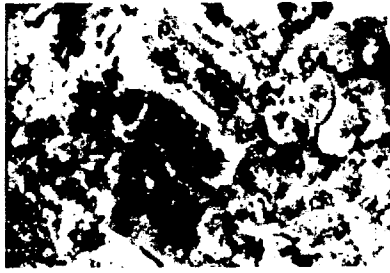
Finally, analysis of the product samples under SEM (Photo 1) showed the transformation of incineration fly ash into new compounds, which explains for the changes in its physical properties. The photographs show that by pressing the fly ash at room temperature (273 K), only changes in its physical structure occur. However, by the incorporation of heat into the compression process, the appearance of new Confirmed by the presence of larger aggregates whose size increases with the temperature increment. The fly ash contains varying amounts of SiO₂, Ca, Na and Cl and it is Thought that the above treatment enhances the hydrothermal reaction of these elements.



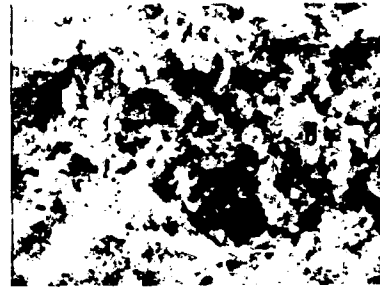
Incineration Fly ash



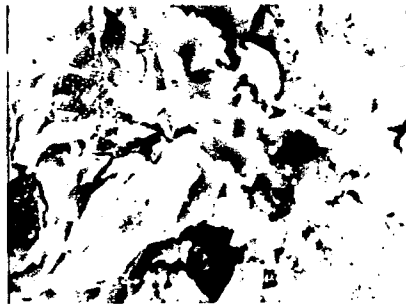
Temp. 273 K



Temp.: 373 K



Temp.423 K



Temp .473K



Temp.523K

Photo 1 SEM photographs of fly ash and fractured surface of consolidated sample at different temperatures.
(Pressure: 20 MPa; Time: 60 min.)

5. Conclusion

The hydrothermal and alkaline hydrothermal methods for heavy metal removal from fly ash were examined with positive results. As compared to normal hydrothermal treatment, the time to extract trapped metals is reduced during alkaline treatment. By hot-press hydrothermal treatment, a consolidated sample with high compression strength of approximately 60 MPa, tensile strength of 6.5-7 MPa and hardness of 20-23 RH15W is produced. Modifications to the solidification procedure such as the mixing of glass to the fly ash resulted to consolidate sample of even higher tensile strength and hardness of 12.5-13.3 MPa and 77-80 RH15W, respectively. With these results, the possibility of employing hydrothermal hot-pressing treatment to convert incineration fly ash into safe building materials is substantiated.

6. References

- [1] H. Katsuura and S. Inoue: PPM, Vol. 4, p. 24-30, (1995) (Japan)
- [2] U. Abe: PPM, Vol. 11, p. 21-26 (1991) (Japan)
- [3] N. Yamazaki: ALPHA, Vol. 5, p. 21-38, 1991 (Japan)
- [4] F. G. Smith: Phys. Geochem., Addison Wesley, p. 335 (1963)
- [5] N. Yamazaki: Ind. Education Soc. Magazine, Vol. 4, No.41, p. 29-33 (1993) (Japan)
- [6] K. Sato et al. :J. Ceram. Soc. Japan, Vol. 106., No3, p. 262 (1997)
- [7] K. Izumi et al.: J. Res. Chichibu Onoda Cement Corp., Vol. 47, No. 130, p. 111-118 (1996) (Japan)