Development of a recycling technique of used glass by conversion to porous materials

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Abstract

The powders of used glass bottles were converted to porous plates by a newly developed technique, which involves two steps, hydrothermal treatment of glass powders at low temperatures and calcination at high temperatures in air for foaming. The ordinary method to prepare porous materials from glass needs vesicants such as calcium carbonate and silicon carbide that decompose at high temperatures to produce gas, and the gas is trapped in softened glass to form pores. The new technique can produce porous materials without any vesicants. Water incorporated into the glass structure by hydrothermal treatments acts as a vesicant. Water diffuses into the glass structure by hydrothermal treatments at low temperatures of around 200°C, and is released as vapor to form pores in the softened glass, when the hydrothermally treated glass is heated at high temperatures over 650°C. Thus, this process gives porous materials with a fine microstructure including closed pores at low temperatures in comparison with the ordinary method. Porous plates (45 x 45 x 3 cm^3 in size) with a bulk density of 0.45g/cm^3 were produced by hydrothermal treatment of the glass powder at 183°C in a large autoclave with an inside volume of 2.5 m^3, followed by calcination at 800°C in a continuous furnace with 18 m in length.
Keywords: recycling of used glass, porous materials, hydrothermal treatment, foaming.

1 Introduction

The material recycling is important for a sustainable development [1, 2]. In Japan, we produced 1,550,000 ton of glass bottles in 2004 [3]. After used, these glass bottles are separated into colors, transparent, brown and others, and recovered for recycling. Their recycled amount was 320,478 ton in 2004 [4]. The used glass bottles were mainly (69%) used as a raw material to produce new bottles, and the remainder (31%) was converted to other materials. The recycling of used glass is attractive for glass manufactures, but they need the used glass with similar composition controlled by severe color sorting and removing contaminants. It is easy to produce new bottles from separated transparent and brown glass bottles, but not from the bottles with mixed colors. Thus, the recycling of colored glass bottles has been received increasing interest. The construction industry has given successful recycling of used glass for heat insulation (fiber glass and light-weight aggregates), aggregates for concrete and asphalt, base and subbase filler materials, and cement constituent [5]. In this study, we developed a new technique to recycle the used glass bottles with mixed colors by conversion to porous plates.

The ordinary method to prepare porous materials from glass needs vesicants such as calcium carbonate and silicon carbide that decompose at high temperatures to produce gas, and the gas is trapped in softened glass to form pores. In this study, we developed a new method by using hydrothermal technique to convert the powder of used glass bottles to porous materials. Figure 1 schematically illustrates the procedures.

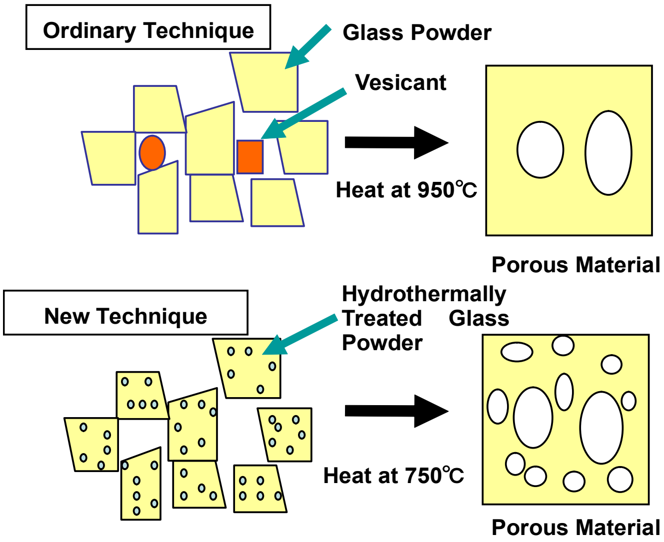

Figure 1: The techniques for preparation of porous materials from glass powders.

The new method involves two steps, hydrothermal treatment of glass powders at low temperatures and calcination at high temperatures in air for foaming. Water incorporated into the glass structure by hydrothermal treatments acts as a vesicant. Water diffuses into glass structure by hydrothermal treatments, and is released as vapor to form pores in the softened glass, when the hydrothermally treated glass is heated at high temperatures. In this method, each glass particle after hydrothermal treatment can foam by heat treatment al low temperature in comparison with the ordinary method.

2 Preparation of porous materials

The starting glass powder was available from Toyo System Plant Co., Ltd., Japan. It was produced by milling of used glass bottles and sieved to be under 590 μm. The powder was placed in a stainless steel box (60 x 40 x 20 cm^3) after mixed with water (20 mass%). The hydrothermal treatment was conducted in an autoclave (Tokai Concrete Ind. Co., LTD.) with inner volume of 2.5 m^3. As shown in Figure 2, 9 boxes filled with the glass powder (320 kg, total glass powder) were hydrothermally treated at once at 183°C for 10 hours in saturated vapor.

Figure 2: Autoclave for hydrothermal treatment.

The hydrothermally treated glass was crashed, sieved again to get a powder to be under 590 μm, and heated in a continuous furnace (Toyo System Plant Co., Ltd., Japan) with width 1 m and length 20 m, as shown in Figure 3. The effective heating region was 18 m and was separated to three zones of which temperature can be independently controlled by oil burners. The hydrothermally treated glass powder was placed on stainless mesh belt and continuously transferred into the heating zones. In this study, the temperature of all heating zones was controlled to be 800°C and the transfer rate was selected to be 50 cm in a minute. Thus, the glass powder was heated at 800°C for 36 minutes. After heated in the continuous furnace, the foamed plate was immediately moved to a furnace at 450°C and kept for 4 hours.

Figure 3: The continuous furnace for heat treatment.

3 Results and discussion

The change of crystalline phases by each procedure was observed by X-ray diffraction (Figure 4). The original glass powder was amorphous (Fig. 4(a)) but a few crystalline phases were formed after the hydrothermal treatment (Fig. 4(b)). They were considered to be hydrated crystalline phases, because they disappeared by heating at high temperatures for a short time. The amount of crystalline phases increased with the increase in hydrothermal reaction temperature and time.

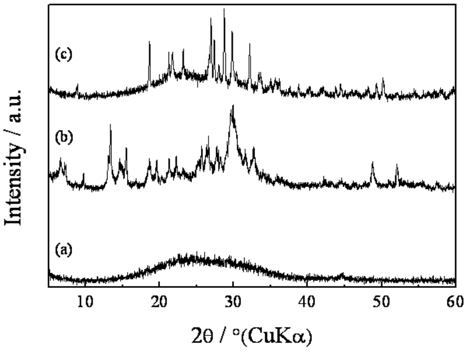

Figure 4: XRD diffraction patterns of the original glass powder (a), hydrothermally treated glass powder at 183°C for 10 hours (b), and plate obtained after heat treatment at 800°C for 36 minutes (c).

It was confirmed by the other experiments that a glass block reacted with water by hydrothermal treatments to form a reaction layer consisting of the crystalline hydrated phases on its surface. The careful observation of the reaction layer showed that the reaction layer produced in water vapor had amorphous material between crystalline part and unreacted part. When the reaction layer formed by the hydrothermal treatment in water vapor was heated in air at 750°C for 30 minutes, pores were formed in the reaction layers. On the other hand, the reaction layers produced in a large amount of water by the hydrothermal treatment never gave pores. It is considered that the hydrothermal treatment in water vapor produces a reaction layer consisting of water diffused glass phase by ion exchange mechanism and the water diffused into the glass structure was released to produce pores when the glass starts to soften at high temperatures.

After heat treatment at 800°C for 36 minutes, the hydrothermally treated glass powders expanded and connected together to form a plate. Though the foaming was observed even at 650°C, the expansion was not enough and the mechanical strength of the plates was very small. In order to get high mechanical strength, heat treatment at high temperatures over 750°C was necessary. When the plate was immediately cooled after the heat treatment, cracks were formed during cooling. After the plate was transferred to the other furnace and kept at 450°C for 4 hours, the plate without cracks was successfully obtained. The plate was consisted of a few crystalline phases together with amorphous material (Fig. 4(c)). The amount of the crystalline phases increased with the increase in heating temperature and time.

The plate had high machinability, so that it was easily shaped into a rectangular parallelepiped. In this study, plates with 45 x 45 x 3 cm^3 in size were produced as shown in Figure 5. The average bulk density of the porous plates obtained in this was 0.45 g/cm^3. The bulk density depended on hydrothermal and heat treatment conditions. It was confirmed by the other experiments that the hydrothermal treatment at 200°C for 6 hours and heat treatment at 750°C for 30 minutes gave low density less than 0.3 g/cm^3.

Figure 5: A shaped porous plate obtained in this study.

The porous plate obtained in this study has a fine microstructure as shown in Figure 6. The pores up to 500 μm were observed in the polished surface of the porous plate. The pore size also depended on the hydrothermal and heat treatment conditions. In general, higher hydrothermal and heat treatment temperature gave larger pores and larger pore diameter distribution. The plates included open and close pores, and floated on water.

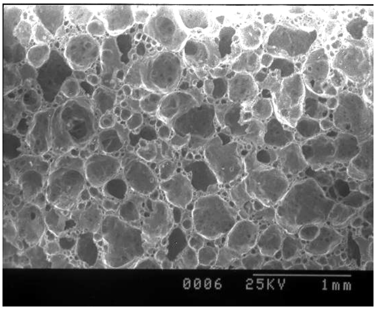

Figure 6: Microstructure of the porous plate.

4 Conclusions

The porous plates were successfully produced from colored used glass bottles by hydrothermal treatment at 183°C for 10 hours, followed by heat treatment at 800°C for 36 minutes. The plates have following properties as average values; bulk density 0.45 g/cm^3, compressive strength 140 kg/cm^2, bending strength 55 kg/cm^2, thermal conductivity 0.18 W/mK, and line expansion coefficient $5.8x10^{-6}$/K. Thus, these plates may be useful as a light weighted board for the void slab system, thermal insulators, sound and water absorbents, floats to form floating islands, and so on.

References

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