

# Development of a New Recycling Process of Fine Aggregate from Waste Concrete Particles Using High- Pressure Carbon Dioxide Solution

Masakazu Nakagawa, Atsushi Iizuka, Minoru Fujii, Kazukiyo Kumagai, Akihiro Yamasaki, Kenji Hikino, Koichi Aoki, Yukio Yanagisawa

## 1. Introduction

Concrete is one of the most commonly used building materials and consists of coarse aggregate (5~40 mm in diameter), fine aggregate (0~5 mm in diameter), cement, and water, with a general mixing ratio of 37, 33, 19, and 11 wt-%. Because cement hydrate consists mainly of calcium silicate hydrate with large amount of calcium hydroxide, it is an alkali substance and known to be damaged by acid rain.

The recycling rate of waste concrete in Japan has rapidly increased in late years along with the enforcement of the “Construction Material Recycling Law” in 2000. The amount of waste concrete generation is about 35 million ton per year of which 96 % is reused mainly as a roadbed (2000). However, the current recycling process has several limitations. The emission of the waste concrete is expected to be in a rapid increasing trend for the next decades and reach as much as 50 million ton per year in 2050<sup>1)</sup> due to the demolition of huge number of buildings constructed in 1970’s, Japan’s high economic growth period, while the demand for roadbed is expected to be on a declining trend in the future with the reduction of public works projects. In addition, shortage of dump yards is a serious problem in Japan; the remaining capacity is estimated to be 13 years (2000). Thus waste concrete could be able to neither recycle nor dispose in near future.

On the other hand, since ensuring high-quality natural aggregate is getting an important issue as resource depletion and regulation for environment protection, development of an effective recycling process of high quality aggregate that can reuse for building material from waste concrete is an urgent issue from both aspects of waste management and resource conservation.

There are several established processes for recovery of aggregate from waste concrete, which can be categorized in two types: mechanical grinding method and heating and grinding method<sup>2)</sup> (Fig. 1). The mechanical grinding method can separate coarse aggregate by crushing and classifying the raw waste concrete but not for fine aggregate; it generates large amount of small particles of waste concrete (SP). On the other hand, the heating and grinding method can recover the both aggregate; however, it is an expensive and energy consuming technique because of heating process. Those methods require waste concrete crushed into concrete block (~40 mm) prior to the processing, which generates SP as much as 20% of the material, and totally, 20 to 76 % of waste concrete is obtained as SP that can not be used as fine aggregate because of high water absorption rate (WA).

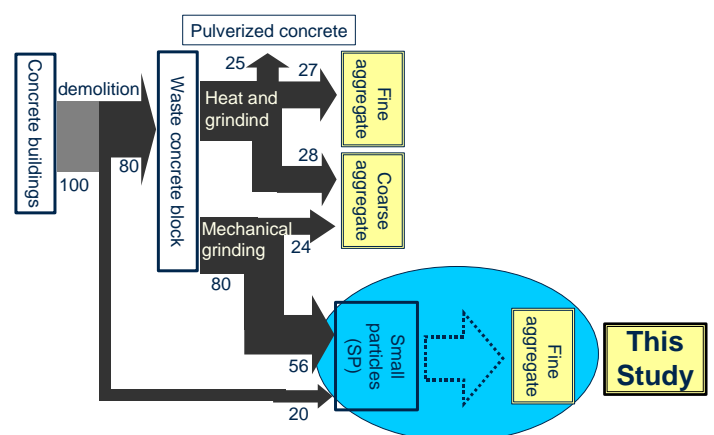


Fig. 1 Existing aggregate recovering process and material flow of waste concrete

In order to recover fine aggregate from SP, cement hydrate, which coats aggregate, need to be removed; however, the recovery processes is still under development because of the difficulty in the separation of these components.

Cement hydrate is alkaline substance, and the calcium constituents are extractable by acid treatment possibly resulting in strength reduction, which is easily crushed and separated by ball mill and screening. In this study, a new type of recycling process of fine aggregate from SP by combing acid treatment and mechanical treatment is investigated. Real sample of SP is treated by high-pressure CO<sub>2</sub> aqueous solution and ball mill as the acid and mechanical treatment, respectively. By using carbon dioxide, a global warming gas required to be reduced, the process have potential to be an environmental friendly method.

## 2. Experimental

### 2.1 Waste concrete sample

Two types of SP (below 5 mm in diameter) are employed in this study. Those are generated in different processes and have different WA. One, obtained by mechanical grinding method aiming at recovery of fine aggregate and having white color with a little amount of cement hydrate coated, is named X having low WA (3.7%). The other one, obtained by crushing waste concrete into concrete block having large amount of cement hydrate and easily generates white powder, is named Y having high WA (15.3%). The rate of calcium content is 11 wt-%, and that for aggregate (>0.15 mm) is calculated to be 59 wt-% for sample Y. Assuming the rate of calcium content for cement hydrate to be 30% and that for aggregate to be 0% does the calculation.

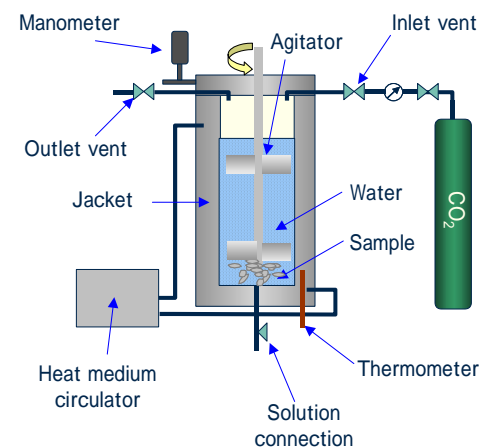
### 2.2 Method

The recycling process is composed of three types of treatments, namely, treating with high-pressure CO<sub>2</sub> aqueous solution, grinding with a ball mill, and classification.

CO<sub>2</sub> treatment is carried out with 1 L agitator as shown in Fig. 1. After 10g of sample and 800 mL of ion-exchange water were set in the vessel, gas phase is displaced and filled with CO<sub>2</sub> to the specific pressure; the liquid phase is agitated at 300 rpm and calcium extraction proceeds. The temperature and pressure are maintained at 323K and 3 MPa, respectively. When extraction carried out for more than 1-hour, the solution and sample is separated by a sieve of 0.15 mm, and fresh water and the sample are set every 1-hour. After the treatment completed, the sample is separated as well and dried at 353K for one night. The solution is collected at a certain interval during extraction, and the calcium concentration is measured by ICP-AES.

Pot of 0.4 L in inner volume and zirconia ball of 10 mm in diameter at filling rate of the ball is 40 % are employed for the ball mill crushing at 100 rpm for 30 min. Particulate is removed either by 0.15 or 0.6 mm sieve afterward.

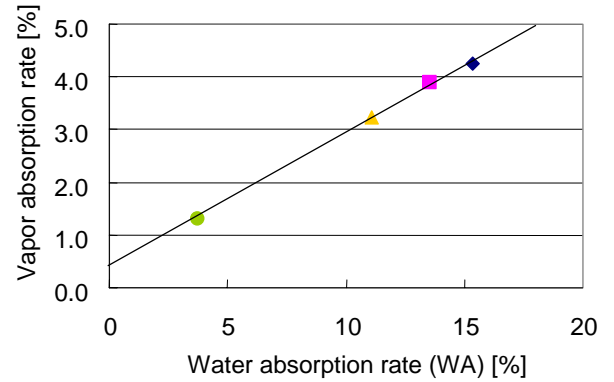
Samples after the treatments removal particulate were defined as “recycled fine aggregate” and the yield is calculated as: weight of recycled aggregate / initial weight (10 g) × 100.



**Fig. 2** High-pressure CO<sub>2</sub> aqueous solution agitator

### 2.3 Measurement of water absorption rate

Japan Industrial Standard (JIS) defines maximum standard of WA to be 3.5% for fine aggregate that can be a material for concrete. Measurement of WA is carried out by a simplified method named “moisture absorption method” using vapor absorption rate as the indicator for WA assumption. Comparison between the vapor absorption rate and WA for several samples shows the linear curve (Fig. 3). Vapor absorption rate is converted to WA by the standard curve.



**Fig. 3** Comparison between vapor absorption rate and water absorption rate  
The four plots correspond to four different samples.

## 3. Result

### 3.1 Result of treatment on sample X

In case of the sample X having rather low WA of 3.7%, single CO<sub>2</sub> treatment reduced WA below JIS standard 3.5% (Table 1). After the treatment, change in particle color occurred from white to black indicating removal of cement hydrate.

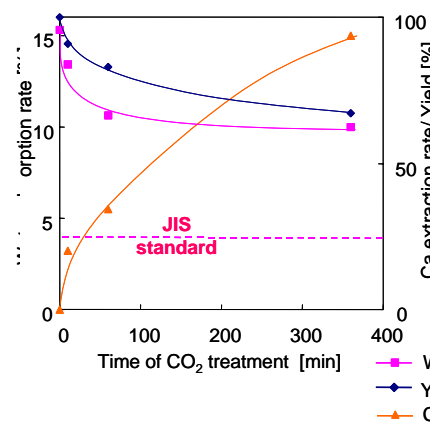
### 3.2 Result on sample Y

6-hour CO<sub>2</sub> treatment extracted more than 90% of calcium content of sample Y while the WA did not fall below 10% (Fig. 4). The surface of the particles was covered by brown substance, which was thought to be a residue of cement hydrate mainly consisted of clay after the calcium extraction. It seemed that the brown substance should be removed for the further reduction of WA.

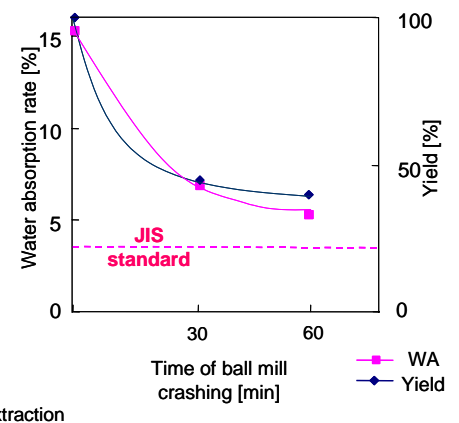
Single ball mill crushing reduced WA to 5.3% after 60 min treatment along with a large drop in the yield (Fig. 5). It seemed that single ball mill crushing may further reduce WA, but is not suitable for the low yield as reported previously<sup>3)</sup>.

**Table 1** Result of CO<sub>2</sub> treatment on sample X

Sample No.	Reaction time [hour]	Water	Particle size
		absorption ratio after treatment [%]	after treatment and filtration [mm]
X-1	1	3.4	0.15-5
X-2	6	3.0	0.15-5



**Fig. 4** Result of CO<sub>2</sub> treatment on sample Y



**Fig. 5** Result of ball mill crushing on sample Y

**Table 2** Combined treatment of CO<sub>2</sub> and ball mill on sample Y  
The reaction time for CO<sub>2</sub> and ball mill is 6 hour and 30 min, respectively

Sample No.	Treatment methods			Water	Particle size
	C: CO <sub>2</sub> solution	B: Ball mill crushing		absorption ratio after treatment [%]	after treatment and filtration [mm]
Y-1	C	B		4.2	0.6-5
Y-2	B	C	B	3.2	0.6-5

Combination of CO<sub>2</sub> and ball mill treatment was carried out (Table 2). Ball mill crushing after CO<sub>2</sub> treatment and followed by sieving at 0.6 mm reduced WA to 4.1% (Y-1). The sieve of 0.6 mm is used to remove crushed cement component thoroughly. Observation of the surface of the obtained particles found white cement hydrate component that indicated incompleteness of CO<sub>2</sub> treatment. In order to avoid incompleteness, sample is pretreated by ball mill, which yielded recycled fine aggregate of 3.2% WA that met JIS standard (Y-2).

## 4. Discussion

### 4.1 CO<sub>2</sub> treatment

The result of CO<sub>2</sub> treatment on sample X shows that a process combining an existing mechanical grinding process and CO<sub>2</sub> treatment could be a new recycling process for fine aggregate. Even so, there are large amounts of SP generation through the existing mechanical grinding process and its material

producing process, so a fine aggregate recovering process for sample Y should be considered.

In case of sample Y, CO<sub>2</sub> treatment extracted more than 90% of calcium that led to the change in color and surface structure observed by SEM (Fig. 5), while the WA did not fall below 10% (Fig. 3) indicating the need of additional mechanical treatment to remove the cement hydrate residue.

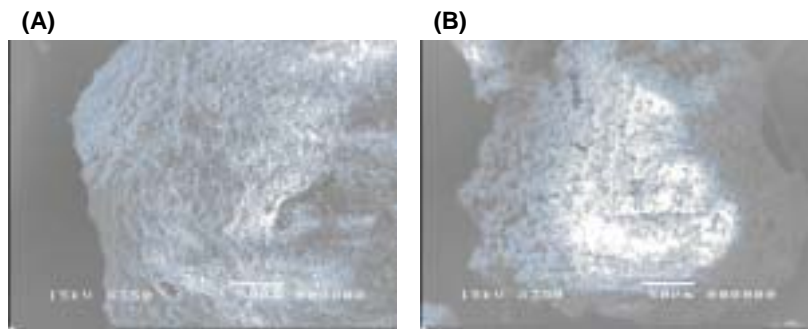
### 4.2 Combining CO<sub>2</sub> treatment and ball mill crushing

Ball mill crushing following CO<sub>2</sub> treatment removed cement hydrate residue and reduced WA considerably (Table 2, Y-1) while some unreacted cement hydrate component was observed on the surface of the particles. This indicates that completion of CO<sub>2</sub> treatment is hardly achieved unless additional treatments are employed, such as mechanical pretreatment as done in this study, cycling process of CO<sub>2</sub> and mechanical treatment, or simultaneous CO<sub>2</sub> and mechanical treatment. Further investigation will be needed.

The power consumption and cost for the recycling process will be estimated to optimize the recycling process based on the experimental studies.

## 5. Conclusion

In this study, it is indicated that two different SP samples can be recycled as fine aggregate that meets JIS standard. Sample X, obtained by mechanical grinding method aiming at recovery of fine aggregate, can be recycled by single CO<sub>2</sub> treatment, and sample Y, having high WA and thus being not considered as suitable material for existing aggregate recovering methods, can be recycled by combined process of CO<sub>2</sub> and ball mill treatment.



**Fig. 5** Surface observation of a particle of sample Y by SEM ( $\times 350$ )  
Fine rolls can be observed before CO<sub>2</sub> treatment (A), while rather rough after CO<sub>2</sub> treatment (B).

## References

- 1) Document issued by Policy Bureau at National Land and Transportation Ministry, 2003
- 2) ISHIKURA Takeshi, et al; Concrete Kogaku (Japanese); Vol. 37, 1999, pp 16-27
- 3) FUMOTO Takayuki, et al; Concrete Kogaku Nenji Ronbun (Japanese); Vol. 22, 2000, pp 1111-1116