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Experimental evaluation of carbon-capture properties of industrial by-products for post-carbon society

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ABSTRACT

In order to contribute to the realization of carbon neutrality in 2050, the authors have focused on the fact that some of the by-products of social activities have the ability to fix carbon dioxide, and have envisioned the creation of a carbon-capture urban environment using these materials. The authors have selected several actual by-products and conducted the constant flow aeration type CO_2 fixation test that we have developed. From the results, the authors propose a prototype of a database of carbon capture capability of CO_2 fixation materials. Furthermore, using this carbon capture capability database, the authors estimate the carbon capture potentials of industrial by-products generated in Japan as some examples. Consequently, this study proposed carbon capture future society concept using CO_2 fixation materials on the basis of the estimation results conducted in this study.

Keywords: Carbon capture, By-products, Carbon dioxide, Fixation, Carbon neutrality

1 INTRODUCTION

Currently, in order to achieve carbon neutrality by 2050 as a plan of the Japanese government, the development and social implementation of technologies for CO_2 capture, storage, and effective utilization based on the plant-intensive method are being vigorously pursued (International Energy Agency, 2021). However, these methods require transportation and storage of CO_2 to remote sites or plants in order to capture, store, and effectively use CO_2 at the CO_2 source, and it also poses the problem of reducing CO_2 emissions in the system operation. The construction of a social system that allows people to reduce CO_2 emissions in their daily social activities, rather than requiring new energy to reduce CO_2 emissions, is considered to be one way to achieve carbon neutrality.

The first author has focused on the fact that cement and concrete by-products contribute to CO_2 fixation to a small extent, based on a survey conducted at a temporary disaster waste storage site after the Great East Japan Earthquake (Komine et al., 2012). In addition, it has been experimentally confirmed that coal ash discharged from coal-fired power plants and iron−steel slag containing calcium have the ability to fix CO_2 (Nohno et al., 2006; Umino et al., 2014; 2017). Based on this background, the authors have considered the possibility of realizing a community-based carbon neutral system, as shown in Figure 1, by utilizing materials with the ability to fix CO_2 among the by-products generated by social activities. In our previous studies, we have developed equipment and methods for the constant flow aeration type CO_2 fixation test that can measure and evaluate the CO_2 fixation capability of materials (Nohno et al., 2006; Umino et al., 2014; 2017; Komine et al., 2023). In this study, the authors attempt to construct a prototype database of CO_2 fixation capability of by-products emitted from social activities with carbon neutrality in mind by utilizing the constant flow aeration type CO_2 fixation test. Furthermore, using this database, we will estimate the carbon capture potentials of some industrial by-products generated in Japan as some examples, and based on the results, we will evaluate the carbon capture effect of urban environments and discuss the concept of a new future society.

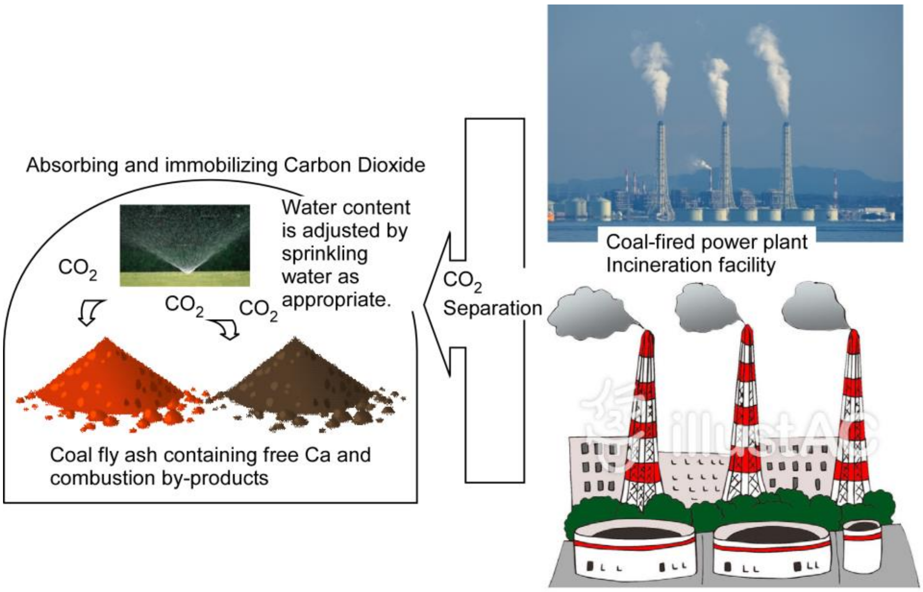


Figure 1. *Conceptual diagram of a carbon neutral facility utilizing coal ash and other CO_2-fixing materials*

2 OUTLINE OF EXPERIMENTS

2.1 Sample

This study used two kinds of soot and dust named as LBB and BB, and paper sludge ash named as PSB-1 and PSB-2. Table 1 shows the fundamental properties of those samples. Soot and dust LBB is dust from a woody biomass boiler, and Soot and dust BB is dust from a biomass boiler that uses wood chips, RPF (waste plastic solid fuel), and waste tires as fuel. PSB-1 and 2 are ashes from a paper sludge boiler that uses wood chips RPF as an auxiliary fuel. The samples are all incinerated ash and discussed on the basis that no organic matter is present. The ignition loss test is currently being carried out to confirm no organic matter in the samples.

Table 1. *Fundamental properties of samples used in this study*

Sample name	Photo	Particle Density	Water Content	pH	Ca Mass Contains Amount	Ca Dissolution Amount
Soot and dust, LBB		2.66 Mg/m^3	5.66%	11.3	10.1 Mass%	214 mg/L
Soot and dust, BB		2.83 Mg/m^3	8.40%	10.1	9.4 Mass%	346 mg/L
Paper sludge ash, PSB-1		2.68 Mg/m^3	0.21%	12.4	18.3 Mass%	216 mg/L
Paper sludge ash, PSB-2			0.06%		29.9 Mass%	366 mg/L

2.2 Experiment apparatus and procedure

This section presents the experimental procedures used for the constant flow aeration type CO_2 fixation test apparatus shown in Fig. 2, and evaluation of the CO_2 fixed in the samples shown in Table 1.

The specimens were prepared in the cylindrical column shown in Fig. 2 and laboratory room air (CO_2 concentration in the range of 400-1000 μL-CO_2/L) was blown into the specimens at a constant flow rate. The air was blown into the specimens through a 2 L gas reservoir filled with distilled water and saturated with water vapor.

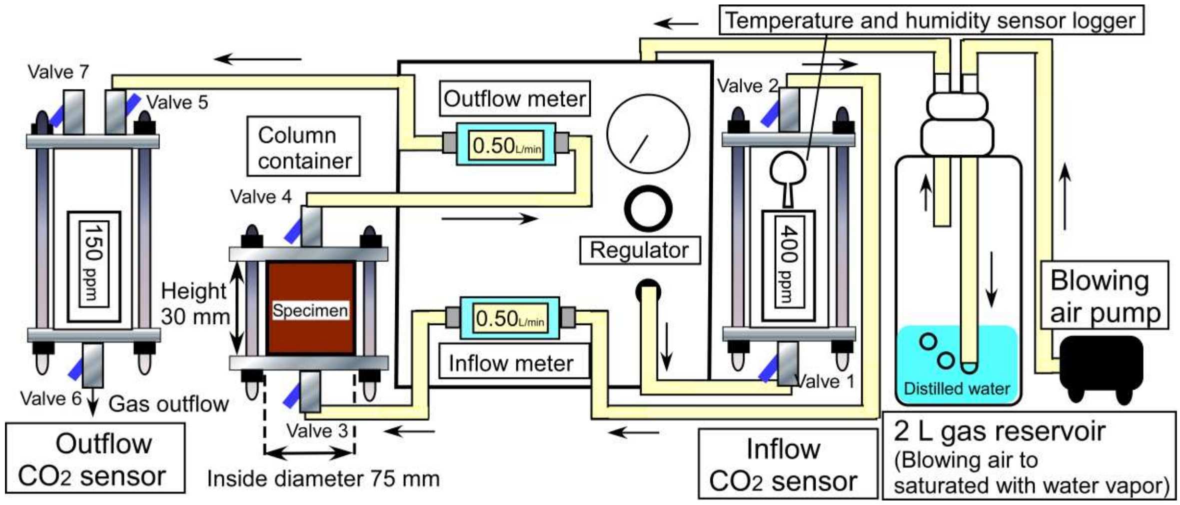


Figure 2. *Constant flow aeration type CO_2 fixation test apparatus*

First, the cylindrical column of the test apparatus shown in Fig. 2 was connected to the pipeline in an empty state, and air was vented using an air pump for approximately 3 hours until the CO_2 concentration in the entire pipeline became almost constant. After that, valves 3 to 5 were closed in sequence, the column was removed from the path, and the specimens were prepared in the column. The specimens were produced by free-falling compaction using samples shown in Table 1 adjusted to the target initial water content of 20%. Height of specimen is 30 mm and inner diameter is 75 mm. When water was added to adjust the water content of the samples, the atmospheric contact time during specimen preparation was in the range of 13 to 15 minutes. Atmospheric contact time was defined as the time between removal of the specimens from the column container, mixing with water, and placement into the column to complete the preparation of the specimens. The samples were prepared as quickly as possible due to concerns that atmospheric moisture could initiate the CO_2 fixation reaction of the samples. During the preparation of the specimens, the air supply to the entire apparatus was continued by connecting the sinflex-tube connected to the valve 3 to the valve 7.

Finally, after the specimen was prepared, the pipeline connected to valve 7 was reconnected to valve 3, and valves 3 through 5 were opened in sequence to resume ventilation of the entire duct. The CO_2 concentration during the test was measured using CO_2 sensors (C2D-W02 wireless CO_2 analyzer manufactured by Udom) installed upstream and downstream of the specimen. The air flow rate was measured by flow meters (Mass flow meter for gases CMS0005) installed upstream and downstream of the specimen.

2.3 Experimental results of the constant flow aeration type CO_2 fixation test

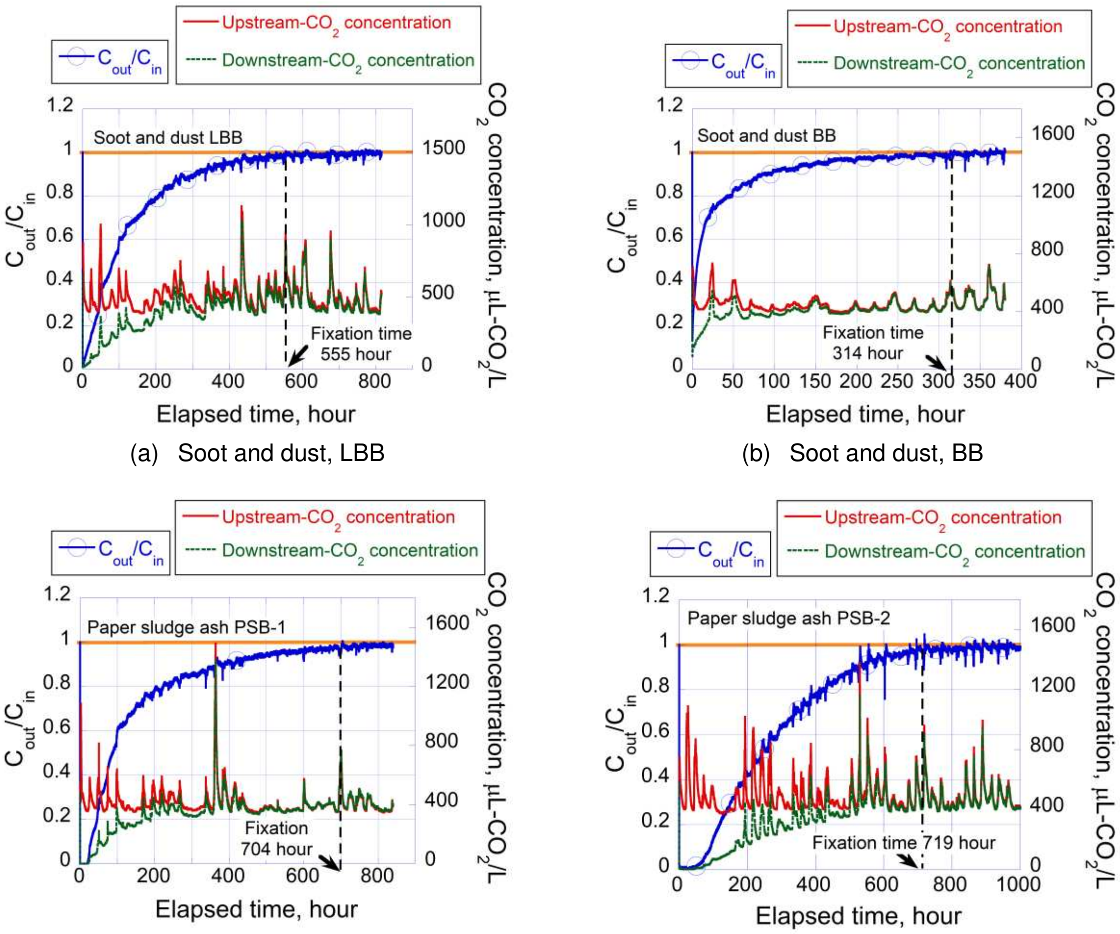
This section describes the experimental results of the constant flow aeration type CO_2 fixation test and the consideration of them.

Table 2 shows the experimental conditions of this CO_2 fixation test that we have conducted. After the specimens were prepared as shown in Fig. 2, the CO_2 concentrations C_{in} (L-CO_2/L) and C_{out} (L-CO_2/L) were measured before (upstream) and after (downstream) the specimens passed through, respectively. The concentration ratio C_{out}/C_{in} was calculated for each elapsed time. As shown in Figure 3, the measured upstream and downstream CO_2 concentrations varied depending on the movement of people in the laboratory and the operation of other equipment, but the CO_2 fixation capacity of the specimens can be evaluated from the concentration ratios.

If C_{out}/C_{in} is less than 1.0 after the specimen is prepared, it means that the downstream CO_2 concentration is lower than the upstream CO_2 concentration, and the specimen can be evaluated as having fixed CO_2 capability. If C_{out}/C_{in} increases with time, it can be inferred that the maximum amount of CO_2 fixation under the experimental conditions has been reached and that no further CO_2 fixation is possible. In other words, by organizing the concentration ratio C_{out}/C_{in} for each elapsed time, the time from the start of the test until C_{out}/C_{in} returns to 1.0 can be defined as the CO_2 fixation time.

Table 2. *Experimental conditions*

Sample	Initial water content	Water content after test	Initial Dry density	Void ratio	Initial CO_2 concentration in upstream	Relative humidity	Test duration
Soot and dust, LBB	17.6%	Not measure	0.45 Mg/m³	4.91	704 μL-CO_2/L	90±5%	816 hour
Soot and dust, BB	20.0%	8.49%	0.51 Mg/m³	4.55	646 μL-CO_2/L		380 hour
Paper sludge ash, PSB-1	14.0%	8.56%	0.33 Mg/m³	7.12	484 μL-CO_2/L		840 hour
Paper sludge ash, PSB-2	10.1%	13.5%	0.41 Mg/m³	5.54	600 μL-CO_2/L		936 hour



(a) Soot and dust, LBB

(b) Soot and dust, BB

(c) Paper sludge ash, PSB-1

(d) Paper sludge ash, PSB-2

Figure 3. *Experimental results of constant flow aeration type CO_2 fixation test apparatus*

Figure 3 shows the change in the CO_2 concentration ratio C_{out}/C_{in} of the specimens prepared from each sample with elapsed time. As a result, it can be inferred that the Soot and dust LBB, the Paper sludge ash PSB-1, the Paper sludge ash PSB-2 have effective CO_2 fixation capability because C_{out}/C_{in} decreased within a certain elapsed time range immediately after the start of the test. For example, in the case of the Soot and dust LBB, C_{out}/C_{in} was 0.0 after the start of the test, and then increased to 1.0 in 555 hours. The degree and duration of the decrease in C_{out}/C_{in} are different for the Paper sludge ashes PSB-1 and PSB-2, but they show the same trend.

3 PROTOTYPE CO_2 FIXATION CAPABILITY DATABASE AND POTENTIAL CARBON CAPTURE CAPABILITY OF SOME INDUSTRIAL BY-PRODUCTS GENERATED IN JAPAN

3.1 Calculation of CO_2 fixation-amount of by-products and prototype CO_2 fixation capability database

This section attempts to construct a CO_2 fixation capability database for each specimen based on the results of the constant flow aeration type CO_2 fixation test described in the previous section.
Based on the CO_2 fixation capability data of the specimens discussed in the previous section, a quantitative evaluation of the amount of CO_2 fixable by each specimen is performed. Figure 4 shows a conceptual diagram of the calculation of the amount of CO_2 fixation by the specimens in the test apparatus shown in Figure 3.

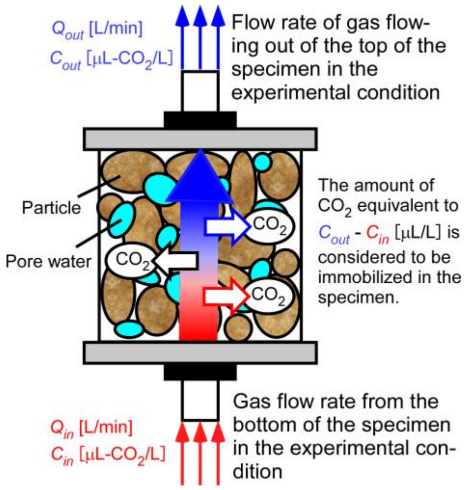


Figure 4. *Schematic diagram of CO_2 fixation in the specimen*

On the basis of the consideration shown in Fig. 4, the amount of CO_2 fixed in the specimen can be calculated from the difference in CO_2 concentration between the upstream and downstream sides based on the following equations (1) and (2) as the equations of state of gas in specimen

$$m_{CO_2}^{SUM} = \int_0^{t_{end}} \left(Q_{in} \cdot \frac{T'}{T} \cdot \frac{p}{p'} \cdot \frac{C_{in} - C_{out}}{10^6} \cdot \frac{M_{CO_2}}{V_m} \right) dt \quad (1)$$

where $m_{CO_2}^{SUM}$ is the mass of CO_2 fixation in the specimen, t_{end} is the time for CO_2 fixation capability to end, T is the temperature of experimental condition, T' is the temperature of standard reference conditions for gases, p is the air pressure of experimental condition, p' is the air pressure of standard reference conditions for gases, C_{in} is the CO_2 concentration of upstream, C_{out} is the CO_2 concentration of downstream, M_{CO_2} is the molecular weight of CO_2 that is 44 g/mol and V_m is the volume per mol of ideal gas that is 22.4 L/mol.

In this experiment, data measurements were taken at intervals of 120 seconds, or 2 minutes. The mass of CO_2 fixed to the specimens during each 2-minutes period was calculated by accumulating in the range $t = 0$ to t_{end} under Eq. (1)

Furthermore, the mass of CO_2 fixation per unit mass of sample is obtained by the following equation (2).

$$m_{CO_2-sample} = \frac{m_{CO_2}^{SUM}}{m_{sample}} \quad (2)$$

where $m_{CO_2-sample}$ is the mass of CO_2 fixation per unit mass of sample, m_{sample} is dry mass of sample in the specimen.

From the experimental results shown in Fig. 3 and the calculation by using the above equations, we can show the prototype CO_2 fixation capability database of by-products as shown in Table 3. This prototype database is attempted to be constructed using experimental data from the four samples shown in Fig. 3 in this study. In addition, carbon capture data of steel slag from a previous study (Umino et al., 2014; 2017) by the first author's group is also shown in Table 4.

It is used as a basis for estimating the carbon capture effect of industrial by-products, which will be described in the next section. It is thought that the CO_2 fixation capability of each sample is affected by grain size of sample's particle, dry density, and water content, etc. In the future, it will be necessary to theorize the relationship between these influencing factors and CO_2 fixation capability.

Table 3. *Prototype CO_2 fixation capability database of by-products in this study* [a]

Sample	Initial water content	Water content after test	Initial Dry density	CO_2 fixation duration	CO_2 fixation amount in specimen	CO_2 fixation amount per unit mass
Soot and dust, LBB	17.6%	Not measure	0.45 Mg/m^3	555 hour	3.31 g	0.055 g-CO_2/g-sample
Soot and dust, BB	20.0%	8.49%	0.51 Mg/m^3	314 hour	0.76 g	0.011 g-CO_2/g-sample
Paper sludge ash, PSB-1	14.0%	8.56%	0.33 Mg/m^3	704 hour	3.45 g	0.079 g-CO_2/g-sample
Paper sludge ash, PSB-2	10.1%	13.5%	0.41 Mg/m^3	719 hour	7.39 g	0.136 g-CO_2/g-sample

[a] Specimen height 30 mm, Specimen inner diameter 75 mm, Air flow rate 0.5 L/min, Target initial water content 20%, Room temperature 25±1℃.

Table 4. *Prototype CO_2 fixation capability database of steel slag in the previous study (Umino et al., 2014; 2017)* [a]

Sample	Initial water content	Water content after test	Initial Dry density	CO_2 fixation duration	CO_2 fixation amount in specimen	CO_2 fixation amount per unit mass
Aged steal slag	8.47%	0.98%	0.97 Mg/m^3	180 hour	3.0 g	0.030 g-CO_2/g-sample
Non-aged steel slag	10.8%	1.12%	0.95 Mg/m^3	180 hour	3.6 g	0.041 g-CO_2/g-sample

[a] Specimen height 20 mm, Specimen inner diameter 75 mm, Air flow rate 0.05 L/min, Room temperature 25±1℃.

3.2 Calculating carbon capture capability of industrial by-products generated in Japan and social image using CO_2 fixation materials for construction

In this section, the authors utilize the carbon capture capability data for the by-products shown in Table 3 to estimate their potential carbon capture effects based on the annual generation of similar by-products produced in Japan. This study selects "steal slag", "paper sludge ash" and "soot and dust" as a by-product of examining the carbon capture effect in Japan.

The statistical data of those by-products is referred from the annual reports of the respective associations such as the Japanese Iron and Steel Slag Association, Japan Paper Association and Ministry of the Environment, Japan. The authors attempt to calculate the carbon capture potential of the by-products by using in combination of the above statistical data and the CO_2 fixation amount per unit mass of the by-products shown in Tables 3 and 4.

Table 5 shows the calculated inferences of carbon capture potentials of the by-products per one year in Japan. From the viewpoint of the annual CO_2 fixation of one ha of forest that is 13.92 Mg/ha-year

(Komine et al., 2023), the results shown in Table 5 are almost CO_2 fixation equivalent to the range of 73,739 to 228,852 ha forests.

Table 5. *Calculated inferences of carbon capture potentials of the by-products per one year in Japan*

Sample	Annual amount (Mg=ton)	CO_2 fixation amount per unit mass (g-CO_2/g-sample)	Carbon capture potential per one year (Mg=ton)	Remarks, Source
Steel slag	12,883,000	0.04	515,320	Japanese Iron and Steel Slag Association, 2022/07
Paper sludge ash (Total amount generated in 2020 year)	4,313,000	0.08	345,040	Japan Paper Association
Soot and dust (High efficiency assumption)	16,609,000	0.14	2,325,260	Ministry of the Environment, Japan
Soot and dust (High efficiency assumption)		0.01	166,090	
Total	33,805,000	-	1,026,450 to 3,185,620	-

Consequently, this study can propose carbon capture future society concept as shown in Fig. 5 using CO_2 fixation materials on the basis of the experimental results and the discussions described as the above.

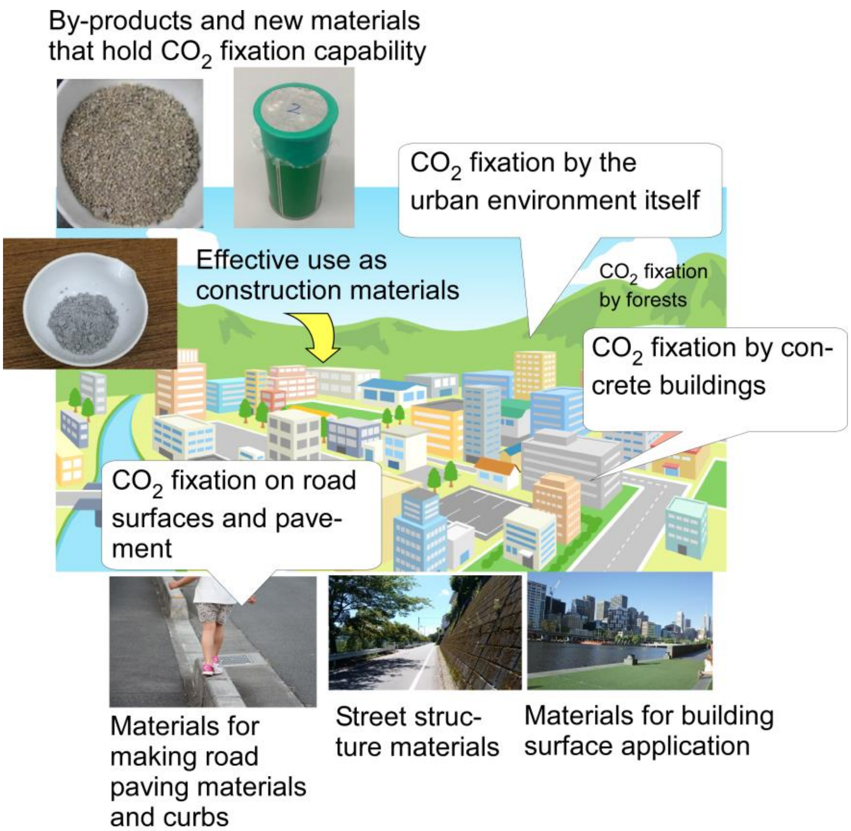


Figure 5. *Carbon capture future society concept using CO_2 fixation materials*

Civil engineering structures are huge and have economies of scale. Even if the CO_2 fixation capability per unit volume and mass is rather small, effective carbon capture effects can be expected at the urban and social environmental levels by utilizing them in large quantities. From this perspective, the field of civil engineering should contribute to the "decarbonization" policy promoted in all of the world.

4 CONCLUSIONS

In this paper, the authors selected four kinds of samples from the viewpoint of the possibility of free calcium retention and appropriate water retention, and proposed a prototype database of CO_2 fixation capability per unit mass by utilizing our originally developed constant flow aeration type CO_2 fixation test. Using this carbon capture capability database, the authors estimate the carbon capture potentials of industrial by-products generated in Japan as some examples. Consequently, this study proposed carbon capture future society concept using CO_2 fixation materials on the basis of the estimation results conducted in this study.

5 ACKNOWLEDGEMENTS

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