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PHYSICAL PROPERTY AND HEAVY METAL LEACHING BEHAVIOR OF CONCRETE MIXED WITH WOODY ASH

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ABSTRACT: In recent years, the woody biomass power generation has attracted a lot of attention since woody biomass is an alternative energy source to fossil fuels. In the future, further implementation of woody biomass power generation is expected, and a large amount of woody ash will be discharged. Several studies have been conducted on the effective use of woody ash for construction materials. However, woody ash has not been practically used because of concerns about the leaching of heavy metals from concrete mixed with woody ash. If the above problem can be solved, a large amount of woody ash is expected to be used. In this study, the fresh property and hardened properties, such as compressive strength and length change were measured to evaluate the feasibility of using concrete mixed with woody ash. Furthermore, a heavy metal leaching test was conducted to assess the environmental impact caused by concrete mixed with woody ash. As the result, the concrete mixed with woody ash has the slump and air content similar to that of normal concrete, the compressive strength meeting the standard durability design strength in Japan, and the shrinkage is similar to that of normal concrete. Additionally, the concentration of heavy metals exceeding the soil environmental quality standards in Japan was partially detected, but the leaching amount of heavy metals from the concrete mixed with woody ash was similar to that from normal concrete.

Keywords: Woody ash, Recycling, Concrete admixture, Heavy metals

1. INTRODUCTION

In recent years, global energy consumption has been increasing due to population growth and economic growth in developing countries. On the other hand, most of the energy resources consumed in the world are fossil fuels, and the reduction of carbon dioxide emissions generated by power generation has become an issue. As the initiative to reduce carbon dioxide emissions, the parties to the United Nations Framework Convention on Climate Change (UNFCCC) have set a long-term goal in the Paris Agreement to hold the increase in the global average temperature to well below 2 ° C above pre-industrial levels and pursuing efforts to limit the temperature increase to 1.5 °C. Woody biomass is a renewable resource and absorbs carbon dioxide through photosynthesis during growth. In addition, woody biomass is an alternative energy resource to fossil fuels. For these reasons, the spread of woody biomass power generation attracts a lot of attention in recent years. In the future, further implementation of woody biomass power generation is expected, and a large amount of woody ash will be discharged. However, in Japan, the reuse of large amount of woody ash. is needed due to the high cost of wood ash disposal and the difficulty of securing new final disposal sites. As reported in [1], most of the woody ash generated is currently landfilled. As shown in [2-7], several studies have been conducted on the effective use of a large amount of woody ash for fertilizer, lime mortar, concrete, and other materials. However, as reported in [2,8], the leaching of heavy metals which are contained in the woody ash is worried due after the service. On the other hand, as shown in [9], hardened cement has the performance of preventing the leaching of heavy metals contained in the waste and it is most commonly used in solidification and stabilization processes. Therefore, if woody ash can be used as a concrete material, a large amount of woody ash can be reused without leaching heavy metals. In addition, substituting woody ash for a portion of cement would also reduce the carbon dioxide emissions associated with cement production. Although the physical and chemical properties of woody ash are influenced by species of tree and tree growing regions, a decrease in the strength of concrete mixed with woody ash has been reported, as shown in [5,7,10]. Therefore, if woody ash is mixed into the concrete, the strength of the concrete may be lower than the strength required for the structure. In this study, slump test, air content test, compressive strength test, and length change measurement were conducted to evaluate the feasibility of using concrete mixed with woody ash by comparing the fresh and hardened properties of normal concrete with those of concrete mixed with woody ash. Additionally, the woody ash used as a concrete material may contain toxic substances, as reported in [11,12], where the content of some heavy metals exceeds the amount of heavy metals in cement or is rarely identified in cement. Furthermore, as reported in [13,14], when the woody ash is mixed with concrete, the unit water content in the concrete must be increased to obtain the same level of workability as that of normal concrete. However, the higher water content in the concrete reduces the degree of setting of the concrete, which may result in an increase in heavy metal leaching from the crushed concrete. In addition, when the woody ash is substituted for a portion of cement, the performance of concrete mixed with woody ash in terms of compressive strength and prevention of heavy metal leaching may be lower than that of normal concrete. To address these issues, it is necessary to verify the heavy metal content in concrete and the amount of heavy metal leaching when various properties of concrete are changed by woody ash admixture. However, few studies have conducted heavy metal leaching tests on concrete mixed with woody ash. In this study, the heavy metal leaching test was conducted in accordance with Notification No. 46 of the Environment Agency in order to examine the relationship between the changes in various properties of concrete due to woody ash mixed into concrete and the leaching amount of heavy metals from concrete.

2. RESEARCH SIGNIFICANT

If woody ash can be used as a concrete material, a large amount of woody ash containing heavy metals can be reused. However, the compressive strength and other properties of concrete mixed with woody ash may be lower than those of normal concrete, and the leaching amount of heavy metals from concrete mixed with woody ash may increase compared to that from normal concrete. In this study, the fresh and hardened properties of concrete mixed with woody ash were measured, and the relationship between these properties and the leaching amount of heavy metals from concrete was also examined.

3. MATERIALS

3.1 Cement, Aggregate and Chemical Admixture

Ordinary Portland cement (3.16 g/cm^3 of density) specified in JIS R 5210 Japanese Industrial Standards was used for cement. Mountain sand with 2.5 mm of maximum grain size and 2.57 g/cm^3 of surface dry density for fine aggregate and crushed stone with 12 mm of average particle size and 2.62 g/cm^3 of surface dry density for coarse aggregate were used, respectively. The air entraining and water reducing agent (AE-WRA) whose main components are a complex of a lignin sulfonic acid compound and a polycarboxylic acid ether was used for chemical admixture.

3.2 Woody Ash

Incinerated ash discharged from woody biomass power plants is classified into bottom ash and fly ash. Most of the ash generated in the incinerator is bottom ash, which comprises about 80 % in total. Therefore, in order to reuse a large amount of woody ash, it is important to examine the feasibility of using concrete mixed with woody bottom ash. In this study, woody bottom ash generated from a woody biomass power plant in Kochi Prefecture, Japan, was used as a concrete admixture. The woody bottom ash contained many coarse particles with 9.5 mm of a maximum grain size. Therefore, in order to obtain a suitable shape for concrete material, woody bottom ash was dried at 100 °C for 24 hours and ground in a ball mill until there were no coarse particles. Fig. 1 shows the woody bottom ash after crushing. The density of the ground bottom ash is 2.47 g/cm^3. Next, the composition of the woody bottom ash was analyzed by XRD (X-ray Diffraction) analyzer. Fig. 2 shows the spectrum of the woody bottom ash analyzed by XRD analysis. As a result of XRD analysis, the composition of the woody bottom ash was 56.5 % of SiO_2, 23.7 % of $CaCO_3$, 11.6 % of Fe_2O_3, 7.9 % of MgO and 0.4 % of CaO by weight. Although the woody ash contained SiO_2, it was not amorphous which promotes the pozzolanic reaction. In addition, in order to confirm the leaching of heavy metals from woody ash, the leaching test was

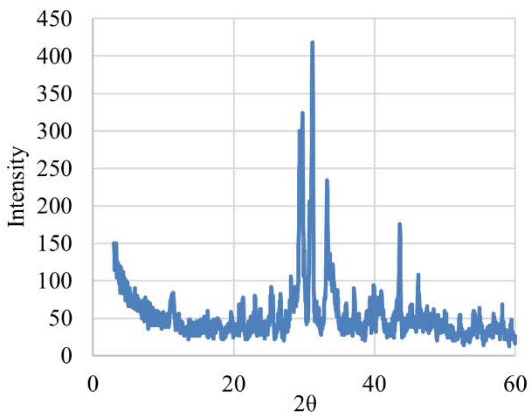

Fig. 1 Spectrum of woody bottom ash by XRD

Table 1 Concentrations of heavy metals leached from woody ash (ppm)

	Woody bottom ash	Soil environmental quality standards
As	0.17	0.01
Cr (VI)	0.41	0.05
F	1.47	0.80

Fig. 2 Woody ash

conducted based on the Environmental Agency Notification No.46. The heavy metals analyzed in the leaching test were arsenic (As), hexavalent chromium (Cr (VI)) and fluorine (F), which are confirmed to be contained in trees in Japan. Cr (VI) and F were analyzed by a monomeric water analyzer based on diphenylcarbazide spectrophotometry, and As was analyzed by ICP-AES. Table 1 shows the concentration of heavy metals leached from woody bottom ash, and all concentrations exceeded the soil environmental quality standard values in Japan.

4. TEST METHOD

4.1 Concrete Mix Design

As reported in [7,15,16], woody ash mixed into the concrete may absorb water and the workability of concrete may decrease. Therefore, when woody ash is mixed into the concrete, it is necessary to increase the unit water volume in concrete in order to achieve the same level of workability as normal concrete. In this test, all concrete mixes were designed with water cement ratio and fine aggregate ratio at 55 % and 43 %, respectively. In addition, the workability of the concreate mixed with woody ash equivalent to that of a normal concrete was ensured by changing the addition quantity of the air entraining and water reducing agent. Table 2 shows the concrete mix proportions in the test. “Blank” and “WBA” represent normal concrete (concrete without woody bottom ash) and concrete mixed with woody bottom ash, respectively. The woody bottom ash mixed into the concrete was woody bottom ash in which heavy metals were leached at concentrations exceeding the soil environmental quality standard values in Japan in the leaching test. In this test, 10 % of the mass of cement used in Blank concrete was replaced with the woody bottom ash. In order to make the workability of the two types of concrete equivalent, the amount of the air entraining and water reducing agent added to the cement mass was 1.0 % for Blank concrete and 1.8 % for WBA concrete. Concrete preparation was carried out based on Japanese Industrial Standard JIS A 1138. The order in which the materials were charged into the mixer was a half amount of aggregate (coarse aggregate and fine aggregate), cement, and the remaining aggregate, to prevent the floatation of powder. The woody bottom ash was mixed into cement in advance and charged into the mixer. After charging the materials into the mixer, the mixer was rotated for 30 seconds to mix the materials. Then, the water added with AE-WRA was poured into the mixer and mixed for 90 seconds.

4.2 Slump Test and Air Content Test

The slump test and air content test were conducted immediately after the fresh concrete was made. The slump test and air content test were conducted based on Japanese Industrial Standards JIS A 1101 and JIS A 1128, respectively. In the slump test, first, the fresh concrete was filled into the slump cone in three times. The fresh concrete in each layer was struck 25 times with a rod, and after the fresh concrete in the third layer was struck, the fresh concrete was made flat to match the top edge

Table 2 Mix proportions of concrete (kg/m^3)

	Water	Cement	Woody Bottom Ash	Fine Aggregate	Coarse Aggregate	AE-WRA
Blank	175	318	0	734	992	3.18
WBA	175	286	32	731	988	5.73

of the slump cone. When the top surface of the fresh concrete was lower than the that of the slump cone, the same concrete sample was added into the slump cone. Next, the slump cone was quickly lifted vertically at a constant speed, and the slump value was measured 0.5 cm unit in the center of the concrete. In the air content test, the air content in fresh concrete was measured based on the air chamber pressure method using a Washington-type air meter. As a method of air content test, fresh concrete was filled into the air content measuring container in the same way as the above-mentioned slump test, and the side of container was tapped with a wooden mallet. The fresh concrete protruding from the container was removed and the top of the concrete was leveled. Next, after the lid was attached to the container, high pressure was applied to air bubbles in the fresh concrete by operating an air meter, which causes volume change. Then, the air content in the concrete sample was measured to the order of 0.1 % based on Boyle's law. In this test, the target slump value and air content value were set at 12 ± 2.5 cm and 4.5 ± 1.5 %, respectively.

4.3 Preparing of Test Specimens for Concrete

A ϕ100 × 200 mm cylindrical specimen was prepared for compressive strength test based on Japanese Industrial Standard JIS A 1132. A cylindrical specimen was removed from the mold, and after that, the specimen was cured in water from the time of specimen preparation until 28 days later. The loading surface (upper surface) of cylindrical specimen was ground with a polishing grinder so that the specimen loading surface (upper surface) became flat. In addition, a 100 × 100 × 400 mm prismatic specimen was prepared for length change measurement. The prismatic specimen was removed from the mold, and then cured in water for 7 days from the time of specimen preparation.

4.4 Compressive Strength Test

After 28 days in water curing of the specimens, the compressive strength test was conducted based on Japanese Industrial Standard JIS A 1108. The compressive strength was calculated by the following Eq. (1).

$$f_c(\mathrm{N/mm^2}) = \frac{P}{\pi d^2/4} \tag{1}$$

where "*fc*" is the compressive strength, "*P*" is the maximum load, and "*d*" is the diameter of the cylindrical specimen measured by a caliper. The compressive strength test was conducted three times for each mix, and the mean value was adopted for the test results.

4.5 Length Change Measurement

The length change of concrete due to drying shrinkage was measured based on the contact gauge method of Japanese Industrial Standard JIS A 1129. As the procedure of the length change measurement, contact chips were affixed to the specimen after curing so that the base length of the chips was 100 mm near the center of the specimen. The contact chips were affixed at two locations on both sides of each side adjacent to the concrete placing surface of the specimen. After that, the concrete specimens were stored in a room at 20 °C and the length change of the concrete specimens was measured. The length change measurement was conducted at 1, 7, 14, 28, and 56 days after the specimens were cured in water. Fig. 3 shows the specimen after contact chips were attached.

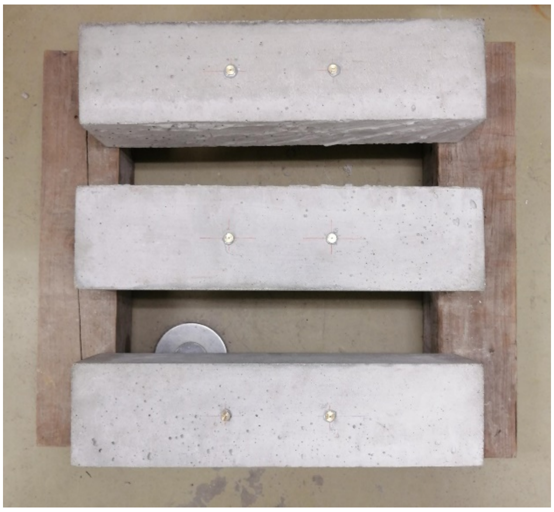

Fig. 3 The specimen after contact chips were attached

4.6 Heavy Metal Leaching Test

As has mentioned above, the concrete has the performance to prevent the leaching of heavy metals. However, there are few studies on the leaching of heavy metals from concrete mixed with woody ash when it is demolished. Therefore, in this study, the heavy metal leaching test on the crushed concrete was conducted based on the Environmental Agency Notification No.46. First, a concrete specimen was crushed with an electric hammer. The crushed one was subjected to sieving with a 2 mm mesh opening, and the one which passed through it was utilized as a sample for the leaching test. Next, 50 mL of distilled water was poured into the centrifuge tube, and 5 g of the sample was also added into the centrifuge tube. It was defined as the specimen in this test and shaken for 6 hours at 200 rpm at 20 °C. After shaking, the specimen was allowed to stand for 30 minutes and then centrifuged at 3000 rpm for 20 minutes. After centrifugation, the supernatant solution of the sample was filtered through a membrane filter with 45 µm of pore size, and the concentration of heavy metals in the filtered solution was analyzed. Cr (VI) and F were analyzed by a monomeric water analyzer based on diphenylcarbazide spectrophotometry, and As was analyzed by ICP-AES. The solution analyzed by ICP-AES was acidified by adding a small amount of nitric acid as a pretreatment. The leaching test was conducted three times, and the mean value of concentrations analyzed was adopted as the test result.

5. RESULTS AND DISCUSSION

5.1 Slump and Air Content of WBA Concrete

Table 3 shows the slump value and the air content value of Blank concrete and WBA concrete. The slump value and the air content value for both fresh concretes were within the target range. From the results, it was found that when 10 % of the mass of cement used in Blank concrete was replaced with the woody ash, the slump value of WBA concrete equivalent to that of Blank concrete could be ensured by adding 0.8 % more the air entraining and water reducing agent than the Blank concrete. Fig. 4 and Fig. 5 show the shapes of Blank concrete and WBA concrete after the slump test, respectively. When the shapes of Blank concrete and WBA concrete were compared after the slump test, there was not any significant difference. Therefore, from the results of the slump test and the air content test, it can be said that WBA concrete has a slump and air content without any problems in construction. These facts are consistent with those described in [7,15,16].

Fig. 4 Shape of Blank concrete

Fig. 5 Shape of WBA concrete

Table 3 Slump value and air content value for Blank and WBA concretes

	Slump value (cm)	Air content value (%)
Blank	12.5	5.5
WBA	14.0	5.5

5.2 Result of Compressive Strength Test

Fig. 6 shows the compressive strengths of Blank concrete and WBA concrete. The compressive strength of WBA concrete decreased by about 19 % compared to that of Blank concrete. The reason why the compressive strength of WBA concrete decreased is considered that the binding force between aggregates decreased by the reduction of cement content in concrete. From the result, it was confirmed that the woody bottom ash did not contribute to the development of compressive strength for the concrete at the age of 28 days. However, the compressive strength of WBA

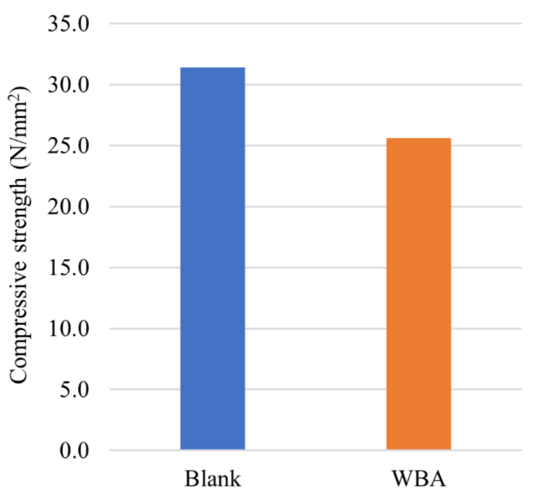

Fig. 6 Result of compressive strength test (28 days)

concrete is 25.6 N/mm^2, which exceeds 24 N/mm^2, one of the standard design strengths applied to concrete structures in Japan. Therefore, even when 10 % of the mass of cement used in normal concrete is replaced with the woody ash, it is possible to use WBA concrete in the same way as normal concrete. According to the report [17], even though the amorphous components were not detected in the woody bottom ash, the long-term strength of the mortar mixed with woody bottom ash increased and exceeded the strength of the normal mortar. Therefore, it is considered that further examination such as the long-term water curing of concrete is needed to obtain more clear knowledge on the strength development of WBA concrete.

5.3 Result of Length Change Measurement

Fig. 7 shows the length changes of Blank concrete and WBA concrete. From Fig. 7, the shrinkage of WBA concrete was about 20 % and 7 % smaller than that of Blank concrete on 14 and 28 days after the start of measurement, respectively. This suggests that the drying shrinkage of concrete can be suppressed by replacing cement used in concrete with woody ash. However, it is necessary to clarify the relationship between the amount of woody ash substitution for cement and drying shrinkage in the future because it is not clear how much cement to be replaced with woody ash is rational and economical.

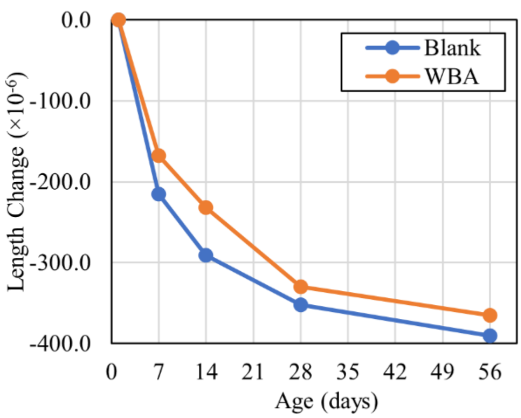

Fig. 7 Result of length change measurement

5.4 Result of Heavy Metal Leaching Test

Table 3 shows the concentrations of As, Cr (VI) and F leached from each concrete. From Table 3, it can be seen that the leaching amount of heavy metals from WBA concrete is equivalent to that from Blank concrete. This fact suggests that there is no significant difference in the amount of heavy metals leached from concrete in which a portion of the cement is substituted with woody ash containing heavy metals, even when the various properties of the concrete are changed, such as a decrease in compressive strength. The soil environmental quality standard values in Japan for As, Cr (VI) and F are 0.01 ppm, 0.05 ppm and 0.80 ppm, respectively. In this test, the concentrations of As and Cr (VI) leached from all concretes sample exceeded the soil environmental quality standard values in Japan. In this study, it is considered that the concentrations of heavy metals exceeding the soil environmental quality standard values in Japan were leached because the leaching test was conducted for the finely crushed concrete. These facts are consistent with those described in [18]. Therefore, when WBA concrete is demolished, it is considered that the treatment to reduce the leaching amount of heavy metals from WBA concrete is needed.

Table 3 Result of heavy metal leaching test (ppm)

	Blank	WBA
As	0.24	0.23
Cr(VI)	0.07	0.05
F	<0.40	<0.40

6. CONCLUSION

In this study, the fresh property and physical properties after hardening, such as strength and length change were measured for normal concrete (Blank) and concrete mixed with woody bottom ash (WBA) to examine the feasibility of using WBA concrete. Furthermore, the heavy metal leaching test was conducted to assess the environmental load caused by WBA concrete.

- From the results of the slump test and the air content test, it was proven that WBA concrete has slump and air content without any problem in construction.
- Although the compressive strength of WBA concrete was slightly lower than that of Blank concrete, it exceeded the standard design strength in Japan.
- The drying shrinkage of concrete was suppressed by replacing cement used in concrete with woody bottom ash.
- The leaching amount of heavy metals from WBA concrete was equivalent to that from Blank concrete. In the meantime, the concentrations of heavy metals exceeding the soil environmental quality standard values in Japan were leached because the leaching test for the finely crushed concrete was conducted.

These findings indicated that the physical properties of WBA concrete after hardening are not significantly different from those of Blank concrete. On the other hand, there is a possibility that the concentrations of heavy metals exceeding the soil environmental quality standard values in Japan are leached from the demolished WBA concrete. Therefore, it can be said that the woody ash can be used as a concrete material by treating the demolished WBA concrete to reduce the leaching amount of heavy metals. However, it is not clear how much cement to be replaced with woody ash is rational and economical. For this reason, it is necessary to clarify the relationship between the amount of woody ash substitution for cement and the properties of WBA concrete, such as the strength development and drying shrinkage, in the future. Additionally, further examination such as a long-term water curing of concrete is needed to obtain more clear knowledge on the strength development of WBA concrete.

7. ACKNOWLEDGMENTS

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