CONTRIBUTION OF STAINLESS-STEEL SLAG TO THE DEVELOPMENT OF STRENGTH FOR SEABED HEDORO

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ABSTRACT

The utilization of industrial wastes as cement replacement material is one of the most desirable methods to solve environmental problems posed by the production of these wastes. Concerning beneficial aspects, the potential uses create new construction materials. There are only a few studies that have been reported on the effective utilization of slag obtained from stainless-steel factory. The objective of this study is, therefore, to illustrate the potential use of a stainless-steel slag as a blended cement material, and to elucidate the contribution of blended slag cement to the development of strength for seabed dredged sludge (hedoro).

The present study investigates hardening effects through the two important strength parameters, unconfined compressive strength (q_u strength) and California bearing ratio (CBR). Evaluation on reaction rate of tricalcium silicate (C_3S) using X-ray diffraction analysis is proposed, and representative data are obtained.

Based on the experimental results, the q_u strength of the hedoro stabilized with slag cement is higher than that of cement-stabilized hedoro in all respects. It is concluded that the slag powders added significantly accelerate the initial reaction rate of C_3S , resulting in an increase in short-term and long-term strength. The development of strength in relation to the hydration mechanism of C_3S is then discussed.

Key words: c. B. R., cement, physico-chemical properties, soft ground, soil stabilization, unconfined compression test, wastes (IGC: D10/K6)

INTRODUCTION

Research reported by many investigators over recent years has confirmed that the industrial wastes such as fly ash and blast furnace slag can be used as blended cement component. Concerning beneficial phenomena, the fine mineral powders accelerate the early strength of

cement (Stein and Stevels, 1964; Ogawa, et al., 1980), reduce heat of hydration (Mather, 1974; Douglas and Zerbino, 1986), and improve resistance to sulphate attack, and to chloride ion penetration when exposed to sulphate as well as chloride solutions (Gjorv and Vennesland, 1979). Several hypotheses concerning the mechanism of cement hydration in the presence

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of pozzolanic admixtures in relation to strength development have been proposed. Kawada and Nemoto (1968) stated that the addition of fly ash accelerated the hydration of cement at an early stage due to the increasing of nucleation sites around cement compounds. Our previous work by scanning electron microscope (SEM) (1989) revealed that fly ash particles in a soil stabilization system behave like spherical cores where reaction products are initially produced, which thus contribute to an increase in ultimate strength. Ogawa, et al. (1980) in the study of hydration in synthesized pure C_3S -pozzolana*** system provided detailed experimental results and proposed two-fold reasons. These combine the concepts of increasing of nucleation sites, and change of Ca^{2+} ionic concentration, to elucidate subsequent mechanism.

The stainless-steel slags are considered as industrial wastes which are unavoidably produced during the manufacturing processes of stainless steel, the increasing amounts of which may create environmental impact in the future due to the limitation of disposal areas. The slag used in this study is of an electric-arc furnace type derived from the Austenitic stainless-steel manufacturing. Annually, the electric-arc furnace slags are produced to the extent of approximately 200,000 tons, however, most of this production has been disposed of in reclamation areas.

This paper concentrates on the study of the potential use of a stainless-steel slag as blended cement material, as well as on the contribution of blended slag cement to the development of strength for seabed hedoro. This was investigated by following the investigation on the changes of strength characteristics, and substantial effect on the reaction rate of C_3S in comparison with those obtained when materials are stabilized with only portland cement. (***) Cement Chemistry notations used in the context are: $C=CaO$; $S=SiO_2$; $A=Al_2O_3$; $H=H_2O$

EXPERIMENTAL PROCEDURE

1) Materials

The sludge material used in this investiga-

tion, generally known as hedoro in Japan, was dredged from the Osaka Bay. The natural moisture content of this so-called seabed hedoro is about 200~300% and may reach 400~500% in the upper layer. Upon visual inspection, the hedoro has a dark-gray color, containing some small fractions of shells, and has a rather strong organic smell. It has high degree of shrinkage, which results in a significant change of bulk volume when dessicated. Further data on physical properties of the soil are given in Table 1. According to the unified soil classification system, the hedoro used can be grouped as an organic clay with high plasticity (OH group).

Table 1. The physical properties of materials used

Property		material	Seabed Hedoro	s-slag
①	Sand fraction (200-75 μm , %)		5.5	23.2
	Silt fraction (75- 5 μm , %)		48.8	76.8
	Clay fraction (less than 5 μm , %)		45.9	—
Initial water content (w_i , %)			100-110	0
②	Liquid limit (LL, %)		110.6	—
	Plastic limit (PL, %)		35.6	—
	Plasticity Index (PI)		75.0	—
Specific gravity (G_s)			2.739	3.216

Note: ① Grain size analysis ② Consistency test

On the other hand, the slag used in this study is of an electric-arc furnace type derived from Austenitic stainless steel manufacturing. The so-called stainless-steel slag (s-slag) contains various compositions, but mainly consist of oxides, such as oxides of calcium (CaO), silicon (SiO_2), and aluminum (Al_2O_3), causing material to possess latent hydraulic properties. Table 2 shows that the major chemical compositions of s-slag are close to those of blast furnace iron slag. The s-slag is composed of

Table 2. The major chemical compositions of electric-arc furnace s-slag in comparison with those of iron blast furnace slag

Type of slag	Chemical Composition (% by dry weight)			
	SiO_2	CaO	MgO	Al_2O_3
s-slag	14.6	35.2	18.6	23.9
iron slag	31	37	8	16

fine-grained particles equivalent to the size of silt grain. When compared with cement grains, the s-slag has lower specific surface area but slightly higher specific gravity; 1875 and 3350 cm²/g and 3.22 and 3.17, respectively.

2) Specimen preparation and Test

Mixing and preparation of specimens was performed in accordance with the Method of making and curing noncompacted-stabilized soil specimens (JSF T821-1988) as prescribed by the Japanese Society of Soil Mechanics and Foundation Engineering. The initial water content for treatment was set to be in the range of 100~110%, which is close to the liquid limit of seabed hedoro. Specimens were prepared by mixing the hedoro with two types of stabilizer; — s-slag blended cement and ordinary portland cement (OPC). The results from a preliminary test revealed that the slag content to be replaced into stabilizer should be 50% or less by weight. A significant reduction of strength, as well as CBR, could be seen when slag proportion was greater than 50%. It was found that a ratio of cement to s-slag equal to 80:20 provided a preferable hardening effect, and thus was used in this study. Mix proportions were 6%, 9%, 12%, and 15% by bulk weight.

From each soil mix, cylindrical specimens of 5 cm diameter by 10 cm long were made for strength tests, and of 15 cm diameter by 12.5 cm long for bearing ratio (CBR) tests. After molding, the specimens were sealed tightly in plastic sheets, followed by aluminum foil, to prevent loss of moisture due to surface evaporation. The former were cured at a room temperature of 20±2°C and 80% relative humidity for periods of 1, 3, 5, 7, 11, 14, and 28 days, while, the latter were cured for periods of 3 days, and then submerged in water for periods of 4 days, before testing, as suggested by the Japan Road Association.

Investigation on C₃S compounds in the mixtures was performed with a Rigaku X-ray diffractometer (using a Cu-target with a Ni-filter and input energy of 40 kV and 20 mA) on consecutive days during the first two weeks, and at the end of four weeks. This study identified C₃S at d-spacings of 2.61~2.62 Å

where strong reflection is prominent and does not overlap with other phases.

RESULT AND DISCUSSION

1) Strength Characteristics of Stabilized Hedoro

The characteristic curves showing the development of strength against curing time are presented in Figs. 1 and 2. Results indicate that the strength increases with the increase

Fig. 1. The strength-time characteristic curves of seabed hedoro stabilized with OPC for various stabilizing levels (HCE) (1 kgf/cm² = 98.1 kN/m²)

Fig. 2. The strength-time characteristic curves of seabed hedoro stabilized with OPC containing stainless-steel slag (80:20) for various stabilizing levels (SCH) (1 kgf/cm² = 98.1 kN/m²)

of stabilizer content and curing period. However, it is apparent that the strength-time characteristic curves for the soils stabilized with OPC (hereafter referred to as HCE mixture) and OPC containing s-slag (SHC mixture) are different. Based on the curve fitting by the method of least squares, the data for SHC and HCE at all stabilizing levels agree with the following equations, respectively;

$$\text{hyperbolic curve: } q_{u1} = \frac{t}{a + b \times t} \quad (1)$$

$$\text{logistic curve : } q_{u2} = \frac{k}{1 + c \times \exp(-dt)} \quad (2)$$

for a, b, c, d , and $k > 0$

To illustrate the contribution of s-slag blended cement on the strength development compared with OPC, we evaluate the ratio of gains in strength of SHC and HCE mixtures (*i. e.*, q_{u1}/q_{u2}) at the same mixing level. As illustrated in Fig. 3, the strength ratios obtained

Fig. 3. The strength ratio between SHC and HCE (q_{u1}/q_{u2}) obtained from; (a) measurement and (b) eq. 1/eq. 2

from measurement conform, and show similar trends, with those calculated from the proposed equations (*i. e.*, eq.1/eq.2). It is obvious that all SHC mixtures (except SHC 6% at longer curing period), obtain higher strength (expressed by the strength ratio greater than 1.0) than HCE mixtures, after 1-day and throughout the course of curing. This superiority is prominent during the early curing periods, particularly at stabilizer content of 9%, however it tends to decline to a constant ratio for longer curing periods. Nevertheless, this can still be considered satisfactory, because strengths obtained

Fig. 4. Comparison between the correlations of CBR- q_u -stabilizer content of hedoro added with OPC and OPC containing s-slag at 7 days ($1 \text{ kgf/cm}^2 = 98.1 \text{ kN/m}^2$)

for SHC mixtures (SHC 9, SHC 12, and SHC 15) are about 10~25 percent higher than HCE mixtures of the same respective mixing level. These results highlight the beneficial effect due to the addition of s-slag into OPC.

Figure 4 illustrates the correlations between CBR and q_u strength, and q_u strength and stabilizer content of the two mixtures for 7-days curing. Similar to the results of compressive strength, the stabilized CBR-value of SHC is greater than that of HCE for the same stabilization level. Obviously, the CBR and q_u strength suggest linear relations with almost the same inclination. In addition, the effectiveness of stabilization is more pronounced when the content of SHC is equal 9% (also see Fig. 3). It is considered that the stabilized hedoro achieves the required properties; — q_u strength and CBR, according to the requirements of the application. We may use stabilized hedoro attaining higher strength as a foundation for small construction such as retaining walls, while, that attaining lower strength, as subgrade for road or fills for embankment. It is most certain that stabilization with s-slag cement requires less stabilizer to achieve required

Table 3. The ratio of 7-day and 14-day strengths to the predicted long-term strength of HCE and SHC mixtures

Mixture	Mixing (%)	Observed Strength (kgf/cm ²)		Predicted Strength (q _u , kgf/cm ²)	Strength Ratio	
		7-day	14-day		q _{u7} /q _u	q _{u14} /q _u
HCE	6	2.60	**	5.56	0.47	—
	9	5.55	8.57	9.58	0.58	0.90
	12	7.48	13.07	13.96	0.54	0.94
	15	10.38	15.44	15.64	0.66	0.99
SHC	6	3.20	4.02	5.08	0.63	0.79
	9	8.47	10.63	13.33	0.64	0.80
	12	9.49	**	20.41	0.46	—
	15	10.46	16.16	23.81	0.44	0.68

Note: ** No observed data

q_{u7}, q_{u14}, and q_u represent compressive strength attained at 7 and 14 days, and predicted strength at long-term, respectively.
(1 kgf/cm² = 98.1 kN/m²)

criteria.

For practical use, the gain in strength at short-term and long-term are equally important. The long-term strength for all mixtures evaluated by using the proposed equations and the strength ratios of the 7-day and 14-day strengths to the predicted long-term strength are summarized in Table 3. We may conclude here that strength of the hedoro rapidly increases in an early age; *i. e.*, from the beginning of mixing to curing periods of 14 days, and the increasing rate gradually decreases after longer curing periods, regardless of the type of stabilizer. When compared with that of HCE mixtures, the increasing rate of strength for SHC is greater, and appears to be more pronounced after 14 days. The evidence of strength development brings to light some essential roles by which the s-slag contributes to strength of seabed hedoro. It is believed that s-slag particles accelerate the hydration rate of C_3S through particular mechanisms. Further discussion is presented to substantiate this assumption.

2) Definition on the Reaction Rate of C_3S Cement Compound and Evaluation by Means of X-ray Diffraction Analysis

Tricalcium silicate (C_3S) is a main compound of cement that influences the properties of portland cement mortar and concrete including stabilized soil. The principal hydration product

of C_3S is a calcium silicate hydrate (CSH) which is involved in providing strength when it become hardened. Therefore, the reaction rate of C_3S directly influences the strength developing mechanism of the cement mixtures, and consequently changes as a function of time at normal hydration. If we assign the rate of change in number of moles of C_3S as $(-dN_c/dt)$, then the reaction rate of C_3S (r_c), defined based on the unit mass of solid in fluid-solid system, can be written as

$$r_c = -\frac{1}{W} \frac{dN_c}{dt} \quad (3)$$

where: W is the mass of stabilizer, and t is time

Using this definition, since the mass of stabilizer (W) is relatively small and constant in the presumed intimate mixture, the reaction rate may thus be empirically assumed as

$$r_c = \frac{-d[N_c/W]}{dt} = \frac{-dC_c}{dt} \quad (4)$$

where: C_c is the concentration of C_3S remaining unreacted in the mixture

To evaluate reaction rate of C_3S , we must obtain and plot the data of the concentration of C_3S against curing time, making a smooth curve-fitting, and thus reaction rate (r_c) is given by a slope on that empirical curve at any time (t_i). Determination of the actual concentration of C_3S seems to be a difficult problem since the stabilized mass contains

varieties of cement compounds and clay minerals. Based on the assumption that the amount of diffracted intensities of C_3S is proportional to a number of moles of C_3S that exists, the concentration of unreacted C_3S can thus be preferably obtained by means of XRD analysis. Figs. 5 and 6 show a number of plots of the diffraction intensity of C_3S against curing time for HCE and SHC mixtures. It is apparent that the reflection decreases exponentially as a function of time. The data is more scattered in SHC mixtures than in HCE mixtures at the same stabilizer content. The non-crystalline components remaining in s-slag is assumed to be the cause of this scattering. Further, the data seems scattered at lower stabilizer content, however, the variation becomes less for the mixtures containing more stabilizer. In most

Fig. 5 (a). The concentration-time curves of C_3S by XRD Analysis of HCE for 6% and 9% stabilizer content

Fig. 5 (b). The concentration-time curves of C_3S by XRD Analysis of HCE for 12% and 15% stabilizer content

Fig. 6 (a). The concentration-time curves of C_3S by XRD Analysis of SHC for 6% and 9% stabilizer content

Fig. 6 (b). The concentration-time curves of C_3S by XRD Analysis of SHC for 12% and 15% stabilizer content

cases, the diffraction intensities significantly change during the first week of curing, and then become almost steady. In addition, the sequence of empirical curves for the same type of stabilizer arranges systematically with the amount of stabilizer added, which thus confirms that the presumed assumption holds. For the mixtures stabilized with different types of stabilizer, it may be incorrect to compare the reactivities through the obtained concentration-time curves directly since the original C_3S contents are different from one another at the beginning. This can be accomplished by determining the rate of depletion of C_3S as a function of time.

3) *Effect of S-Slag on the Reaction Rate of C_3S and Subsequent Strength Increasing Mechanism*

Table 4. Comparison of Reaction Rates of C_3S

Number of day after mixing	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Type of Stabilizer	Stabilizer content (%)	Reaction Rate of C_3S (r_c , cps/day)												
Ordinary Portland Cement (OPC)	6	16.50	10.75	8.00	5.67	3.75	3.11	2.36	2.00	1.31	1.00	0.90	0.79	0.79
	9	23.77	19.60	12.80	9.33	6.75	4.89	3.50	2.91	2.50	2.00	1.63	1.06	0.54
	12	26.88	19.00	15.21	11.17	8.14	6.38	5.11	4.20	3.45	3.16	2.54	2.14	1.50
	15	30.50	19.50	15.25	12.00	11.75	8.00	6.50	5.06	4.83	3.13	2.93	2.00	1.50
OPC(80%) + S-Slag (20%)	6	16.25	11.00	6.25	3.68	3.00	2.25	1.75	1.33	1.00	0.55	0.38	0.21	0.21
	9	28.50	18.51	10.50	8.11	6.38	4.13	3.50	2.50	1.63	1.00	0.44	0.36	0.36
	12	32.57	17.37	8.43	6.45	5.00	4.02	3.33	2.83	2.25	1.25	1.15	0.75	0.39
	15	36.78	19.00	11.50	7.00	5.00	4.38	3.33	2.75	2.42	2.00	1.92	1.00	0.83

The analysis of the C_3S reaction rate (r_c) for all mixtures are summarized in Table 4. Further, the comparison between these reaction rates for the same stabilizer content in relation to the development of strength are illustrated in Figs. 7 (a)~(d). For both mixtures, the calculated r_c increases as the stabilizer content is increased, but decreases exponentially as the curing time increases. The results as shown in Fig. 7 (a) indicates that there is no significant difference in the dissolution rates of C_3S between two mixtures when the stabilizer content is relatively small. This is, therefore, analogous to the results from strength test.

For the mixtures containing proportionately

Fig. 7 (a). Comparison between the strength development, in correlation to the C_3S reaction rate of the HCE and SHC, for stabilizer content equals 6% ($1 \text{ kgf/cm}^2 = 98.1 \text{ kN/m}^2$)

Fig. 7 (b). Comparison between the strength development, in correlation to the C_3S reaction rate of the HCE and SHC, for stabilizer content equals 9% ($1 \text{ kgf/cm}^2 = 98.1 \text{ kN/m}^2$)

more stabilizer, the distinction between reaction rate of C_3S -time curves becomes more evident, [see Fig. 7 (b)]. It is considered, as shown in Fig. 7 (c) and (d), that the C_3S reaction rates for SHC mixtures can be illustrated by two separate functions of time, designated as a linearly declining function when curing time is less than two days, and as an exponentially declining function for later times. For HCE mixtures, it is more consistent to express it by the latter function only. Obviously, the reaction rates of C_3S for SHC mixtures are greater than those of HCE mixtures in the beginning course of reaction. However, the reactivities seem to be more pronounced for HCE mixtures when curing time is greater than 2 days. With

Fig. 7 (c). Comparison between the strength development, in correlation to the C_3S reaction rate of the HCE and SHC, for stabilizer content equals 12% ($1 \text{ kgf/cm}^2 = 98.1 \text{ kN/m}^2$)

Fig. 7 (d). Comparison between the strength development, in correlation to the C_3S reaction rate of the HCE and SHC, for stabilizer content equals 15% ($1 \text{ kgf/cm}^2 = 98.1 \text{ kN/m}^2$)

a more general expression, this means that the C_3S in SHC mixtures initially reacts more vigorously with water in the presence of slag particles, attributing to a significant low C_3S concentration in the mixtures. Therefore, the later reaction rates incline close to steady state earlier than in the HCE mixtures.

The results indicate that the s-slag accelerates the hydration of C_3S compound at an earlier age. Consequently, this influences the degree of crystallinity of calcium silicate

Fig. 8. The strength attained in relation to X-ray intensity of CSH reaction product observed at 3-day curings ($1 \text{ kgf/cm}^2 = 98.1 \text{ kN/m}^2$)

hydrate phases (CSH), which are the main reaction products that contribute to strength of stabilized soil. As obvious from Fig. 8, the attained strength increases in correspondence with the X-ray intensities of CSH products that form. In essence, the higher reflections are obtained from SHC samples.

To explain the roles of stainless-steel slag on the reaction mechanisms of C_3S and its contribution to the strength development mechanism in the blended slag cement-stabilized soil system, hypotheses are developed using the ideal schematic diagrams as shown in Figs. 9.

Concerning primary significance, a slag particle may arrange itself between the particles of C_3S . Such insertion considerably dissipates the C_3S flocs, resulting in an increase of their reacting surfaces, [see Fig. 9 (a)]. The concentration of Ca^{2+} ions and pH in the mixed pastes are significantly increased due to the initial hydration on the surfaces of cementitious compounds including slag. Accordingly, the thin hydrate films of calcium silicate (symbolized as h) are produced protecting the unreacting grains of C_3S and slag. An increase of pH thus leads to the dissolution of silicate and

Fig. 9. Ideal schematic diagrams illustrating the reaction mechanisms of C_3S in the presence of s-slag in relation to the development phases of strength, where c is clay particle, m is mineral, h is hydrate film, and Et is ettringite

aluminate from clay minerals and other minerals (symbolized as c and m) into a pore solution, resulting in relatively high-ions concentration in the so-called gelatinous phase. As a result, there is no significant change in the engineering properties during this stage.

It is considered that the reactions occur between lime and slag, and lime and some anions such as Cl^- , regarded as a pozzolanic reaction and the so-called secondary reaction, are responsible for the reduction of Ca^{2+} ions in the pore solution. This indicates that slag particle plays an essential role on the dissolution of C_3S by repetitively stimulating further reaction

of the C_3S grains, which, therefore, leads to the collapse of the thin protective hydrate films that cover the C_3S and slag surfaces. During the hardening phase, as illustrated in Fig. 9 (b), the C_3S and the slag particles are both acting like precipitation cores where hydration products are gradually formed. The hardening reaction products such as the hydrates of calcium silicate and aluminate and ettringite produced, then replace the spaces which are previously filled with pore water, and combine the mineral compositions together. Especially, it is believed that the formation of ettringite ($3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$) causes a significant decrease in moisture content by combining large amount of water in its crystals, leading to a development of an early strength in stabilized soil (Kamon and Nontananandh, 1989). The hydration products gradually strengthen and become more rigid as the reaction progresses.

Consequently, the increase of strength is attributed to the changes of the properties of the hardened matrix that form among particles from a loose structure to a denser one, [see Fig. 9 (c)]. The porosity and moisture content of the mixture are greatly reduced, and thus are assumed to be the cause of the intermittence of reaction at longer curing periods. Based on the testing results, it is believed that a relatively strong matrix is constructed within the first two weeks.

The previous work (Kamon, et al., 1989) revealed that the formation of reaction products such as ettringite significantly improved the leachate characteristics of s-slag blended cement stabilized soil by combining metallic ion such as Fe^{3+} and/or heavy metal such as Cr^{3+} , and fixing them in its crystal. This confirmed another interesting role of ettringite in stabilized soil, as well as confirming the potential uses of stainless-steel slag and stabilized hedoro for construction materials.

CONCLUSIONS

Based on the test results, it is concluded that the stainless-steel slag can be potentially used as a cement replacement material when the slag content is 20 percent or less. Stabilized

soils appear satisfactory for extensive ranges or purposes due to the preferable strength obtained. In conclusion, initial water content, additive-filling effect, stabilizer concentration, condition of ambience, and time for curing are important factors that control the degree of stabilization. It was also found that the strength developing mechanism of the stabilized hedoro was substantially influenced by the initial reaction rate of the tricalcium silicate (C_3S).

The explanation of the strength development mechanism of soil-stabilizer mixture is illustrated simply by consideration of the changes of phases during the course of reaction. The slag particle added, considerably affects the reaction rate of C_3S , and provides substantial strength development at short- and long-term.

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