MECHANICAL PROPERTIES OF SULFUR CONCRETE

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Mechanical Properties of Sulfur Concrete

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Abstract

Fossil fuel consumption is rapidly increasing in the world, and so is the amount of sulfur that is generated as by-product of the industrial refining process. Since sulfur is expected to increase continuously in the future, the huge cost of waste disposal will be required if there is no counterplan. As a result, using sulfur as construction materials such as asphalt and concrete was considered. However, sulfur concrete made with unmodified sulfur has limitation for practical use because it has inferior properties such as poor resistance to water and significantly more brittle than traditional concrete. In order to overcome these drawbacks, Modified sulfur was developed. Sulfur concrete using modified sulfur binders shows excellent durability in high acid or salt concentrations.

This study investigated the mechanical and durability properties of sulfur concrete made with modified sulfur binder instead of Portland cement. Preliminary tests were performed to evaluate effects of the maximum size of coarse aggregate, the proportion of binder, aggregate, and modified sulfur binder, the replacement rate of SPB by fly ash on the workability, strength, and elastic modulus. 7 cases of specimens were tested with different types and sizes of aggregate, and various proportions of fly ash and SPB, to find several optimal mix proportions to minimize the amount of SPB with preserving workability. Compressive and tensile splitting strength tests were performed and elastic modulus of sulfur concrete was measured. For the specimens made with the maximum size of 19 mm, 13 mm, and 25 mm coarse aggregate, the average compressive strength was 76, 53, and 50 MPa, respectively. When the proportion of fly ash was increased to 5, 12, and 15% as a replacement of SPB, the compressive strength of sulfur concrete showed 76, 83, and 72 MPa, respectively. Therefore, the case with 19 mm coarse aggregate and 15% fly ash (by weight) presented the best mechanical properties.

Based on the results of the preliminary tests, three types of specimens were tested to investigate the properties of sulfur concrete in severe environment, such as freezing and thawing resistance, coefficient of thermal expansion, and chemical resistance. In resistance test in acid and salt solution, The F case showed the most significant strength reductions, while the R case presented the smallest strength reductions in the three different solutions after immersion of 60 days. The average of the measured coefficients of thermal expansion of sulfur concretes is 15.26×10^{-6}/˚C. This value is bigger than that of Portland cement concrete (10.0~13.0×10^{-6}/˚C). The tested sulfur concretes presented high resistance to freezing and thawing. S- and F-type specimens made with natural aggregates had 84.6% of relative dynamic elastic modulus after 300 cycles of temperature change. However, R-type specimens made with recycled aggregates showed 77.6%.

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CHAPTER 1. INTRODUCTION AND BACKGROUND

Fossil fuel consumption is rapidly increasing in the world, and so is the amount of sulfur that is generated as by-product of the industrial refining process. [**1**] In 2007, the global production of sulfur was approximately 1,000 million tons. By 2009, 120 million tons of sulfur was generated in Korea. 90% of the sulfur generated in Korea is the by-product from the desulfurization of oil refining process. Even though some of sulfur is consumed as industrial chemical, most is exported at a low cost due to a lack of domestic demand. [**2**] Sulfur is expected to increase continuously in the future. The huge cost of waste disposal will be required if there is no counterplan. Increased sulfur emissions have led many researchers to look into how this largely unwanted chemical/material could be used. As a result, using sulfur as construction materials such as asphalt and concrete was considered. [**3**] On the other, climate change, which is called as global warming, is one of the most critical global issues that have potential to jeopardize the sustainability of human society. Among many causes, the construction industry is responsible for a major portion of green-house gas emission. For example, the production process of cement itself yields approximately 7% of the total CO2 emission worldwide. [**4**] Therefore, sulfur is an environmentally promising material for concrete to replace cement as a binder.

However, sulfur concrete made with unmodified sulfur has limitation for practical use because it has inferior properties such as poor resistance to water and significantly more brittle than traditional concrete. [**5**] In order to overcome these drawbacks, Modified sulfur was developed. Modified sulfur was showed improvement. Sulfur concrete using modified sulfur binders shows excellent durability in high acid or salt concentrations. Sulfur concrete achieves 70 to 80% of the maximum compressive strength within 24 hours. [**6**]

On this wise, structural damage of fertilizer and metal refining industries by acid and chemicals can be reduced by using sulfur concrete which has exceptional physical characteristics and resistance to attack by a wide range of acids and corrosive materials. [7] Despite the potential possibility of the development for sulfur concrete, insufficient research has been done. This study investigated the mechanical and durability properties of sulfur concrete made with modified sulfur binder (SPB) instead of Portland cement. Preliminary tests were performed to evaluate effects of the maximum size of coarse aggregate, the proportion of binder, aggregate, and modified sulfur binder, the replacement rate of SPB by fly ash on the workability, strength, and elastic modulus. Based on the results of the preliminary tests, three types of specimens were tested to investigate the mechanical and durability properties of sulfur concrete such as freezing and thawing resistance, coefficient of thermal expansion, and chemical resistance.

CHAPTER 2. LITERATURE REVIEW

Sufficient sulfur is recovered as a byproduct at petroleum refineries and natural gas processing plants. The amount of sulfur which is presently being produced is more than the demand for sulfur worldwide. Huge quantities of sulfur is consumed in many industries, but not as much as in produced. [8] Although not widely used currently, sulfur construction material can offer improvements over more traditional materials in specific applications. Sulfur construction materials can include sulfur concrete and sulfur-extended asphalt pavements. [8]

When unmodified sulfur and aggregate are mixed in high temperature as sulfur concrete, the sulfur binder crystallized from the liquid state as monoclinic sulfur (S_β) at 119˚C. On cooling to below 114 ˚C, S_β starts to transform to orthorhombic sulfur (S_α), which is stable form of sulfur at ambient temperatures. By reacting sulfur with an unsaturated hydrocarbon, dicyclopentadiene (DCPD), stable sulfur cements were developed by the formation of long-chain polymeric polysulfides. [9]

Loov et al. [10] compared sulfur concrete with a traditional Portland cement concrete. The cost of the materials for producing sulfur concrete may be expected to exceed Portland cement concrete in areas with high sulfur costs. However even with a small cost differential, sulfur concrete warrants consideration where its special properties may be advantageous compared to Portland cement concrete. Mohamed and Gamal [8] also mentioned that comparison of properties of sulfur concrete with those of Portland cement concrete researched by STARcrete™. The results of comparisons are shown in Table 1.

Table 1: Sulfur Concrete Properties compared with Portland Cement Concrete [8]

Property	Compared with 34.5MPa Portland cement concrete	Test laboratory
Abrasion resistance	Much greater	Daw Chemical, Texas Division, Freeport, USA
Bond strength to concrete	Much greater	Daw Chemical, Texas Division, Freeport, USA
Bond strength to reinforcing steel	Greater	R. M. Hardy & Associates, Canada
Coefficient of linear expansion	Equivalent	R. M. Hardy & Associates, Canada
Compressive creep	Less	R. M. Hardy & Associates, Canada
Compressive strength	Greater	EBA Engineering Consultants, Canada J. A. Smith & Associates, Canada Bernard & Hoggan Engineering, Canada
Corrosion resistance	Much greater	Sulfur Innovations, Canada Mellon Institute, Pittsburgh, USA

Durability under thermal cycling	Equivalent or higher	Ontario Research Foundation, Canada Sulfur Innovations, Canada
Fatigue resistance	Much greater	Iowa State University, Ames, USA
Fire resistance	Slightly less	Sulfur Innovations, Canada Wamock Hersey, Vancouver, Canada
Flexural strength	Greater	EBA Engineering Consultants, Canada J. A. Smith & Associates, Canada
Modulus of elasticity	Greater	R. M. Hardy & Associates, Canada
Splitting tensile strength	Greater	R. M. Hardy & Associates, Canada J. A. Smith & Associates, Canada
Thermal conductivity	Less	Ontario Research Foundation, Canada
Water permeability	Much less	Chemical & Geological Laboratories, Calgary, Canada

* STARcrete^{TM} Technology, 2000

CHAPTER 3. TEST METHODS

I. Manufacturing Process of Specimens

Sulfur concrete specimens were fabricated using the proposed method by ACI 548.2R-93 [**6**] "Guide for Mixing and Placing Sulfur Concrete in Construction." Figure 1 shows the mixing equipment used to fabricate specimens in this study. Simple descriptions of the mixing procedures are presented in Figure 2. At first, coarse and fine aggregates preheated in an oven at 180 ˚C for 6 hours were added to the container that was preheated up to 130˚C by a heating jacket. After 1 minute of dry mixing, modified sulfur binder and fly ash were also poured into the container. Mixing continued until the sulfur binder liquefied, and for an additional10 min after the liquefaction. Then, sulfur concrete was cast and compacted. The produced sulfur concretes were de-molded after 3 days and kept in room temperature (20-25 ˚C) and humidity.

Figure 1: Mixing equipment

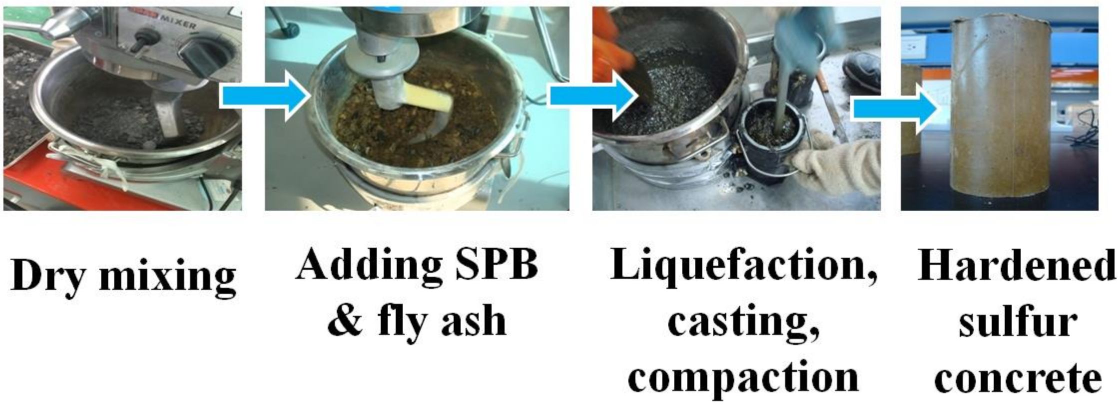


Figure 2: Mixing procedure

II. Strength and Elastic Modulus Tests

Cylindrical specimens used for compressive and splitting tensile strength tests were 100 mm in diameter and 200 mm in height, fabricated according to ACI 548.2R-93. [6] The compressive strength tests were conducted following ASTM C39. The splitting tensile strength tests followed ASTM C496. In order to measure the stress-strain relationship of sulfur concrete, three compressometers that have a 100mm gage length were used. The 1500kN SATEC^{TM} Series 1500HDX hydraulic universal testing machine shown in Figure 3 was used to perform strength test and measure the elastic modulus.

The modulus of elasticity for concrete under uniaxial compression is taken as the slope of the stress-strain curve. Since the stress-strain relationship for concrete is nonlinear, three different methods are typically used to calculate the modulus. Firstly, the tangent elastic modulus at certain point is given by the slope of the line drawn tangent to the stress-stain curve at any point. The secant elastic modulus is given by the slope of the line drawn from the origin to the point on the curve corresponding to 40% of the maximum load. Lastly, the chord elastic modulus is given by the slope of the line that is from the point representing a strain of 50×10^{-6} mm/mm to the point corresponding to 40% of the maximum load. [**11**] In this study, the secant elastic modulus was calculated and used to compare the elastic modulus of sulfur concrete.

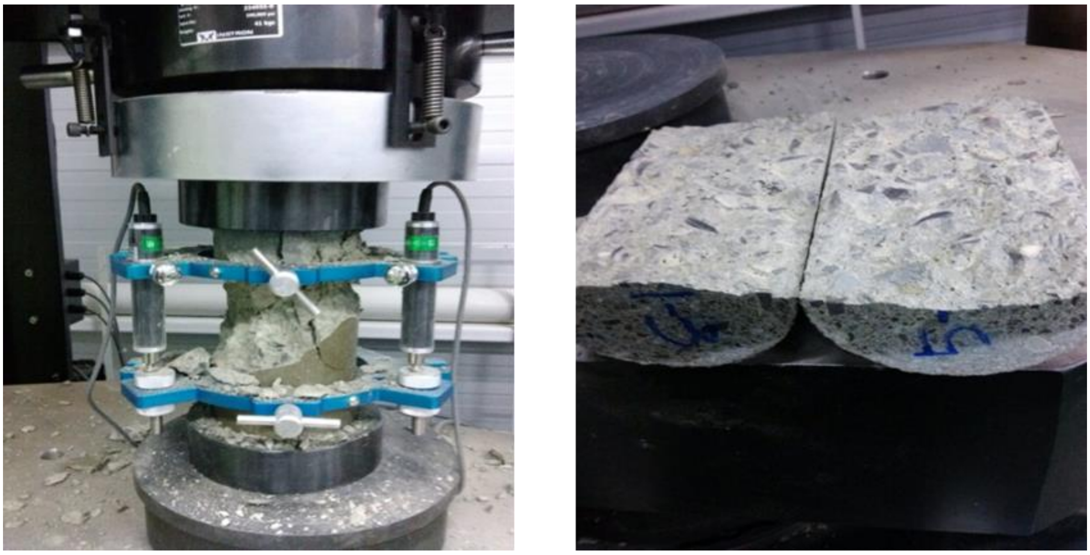


Figure 3: Compressive and splitting tensile strength tests

III. Chemical Resistance Tests

In order to evaluate the chemical resistance of sulfur concrete, the cylindrical specimens were immersed in three different aggressive chemical environments: 10% HCl solution, 20% H_2SO_4 solution, and 3% NaCl solution. The resulting destruction of sulfur concrete during 60 days was observed by measuring the mass periodically on digital laboratory scale and compared with initial mass. Also, the compressive strength after 60 days immersion was measure. Before determining the mass change and compressive strength, the specimens were removed from the chemical solution, washed and dried in an oven at 105℃. Three specimens were used to measure the compressive strength of sulfur concrete before immersion. The mass change and compressive strength of three specimens were also measured after immersion for each case.

IV. Coefficient of Thermal Expansion Tests

The expansion and contraction of sulfur concrete due to temperature changes can impact the durability. In this study, the AASHTO Designation: TP 60-00 (2007) was followed to evaluate the coefficient of thermal expansion of sulfur concrete. First, the specimen was soaked in water for 2 days and the length of the specime was measured. The specimen was placed in the support frame which was submerged in the water tank. The temperature of the water tank was adjusted from 10 ℃ to 50 ℃. The change of the length due to the temperature was recorded by LVDT. [**12**] The linear expansion of a specimen was measured at a heating rate of 0.2 ℃/min. The equipment for measuring the coefficient of thermal expansion is shown in Figure 4.

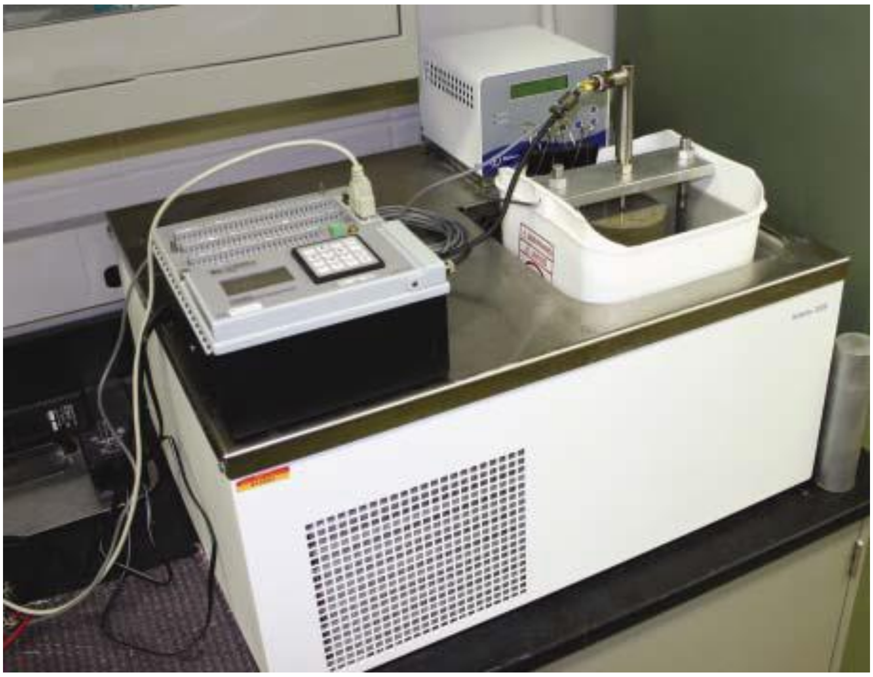


Figure 4: Testing equipment for the coefficient of thermal expansion

V. Resistance of Freezing and Thawing Tests

Tests for the freezing and thawing resistance of sulfur concrete were carried out in accordance with ASTM C 666 procedure B on 100×100×400 mm prismatic specimens. Three specimens were tested for each mixture case at the same time. The relative dynamic modulus of elasticity of the specimen was measured at each 100 cycles. One cycle took 4 hours and was repeated for 300 times with the temperature range from 4 to -18 ℃. The test was terminated when the number of cycle reached 300 cycles or the dynamic modulus of elasticity had decreased below 60% of the initial value before 300 cycles.

CHAPTER 4. PRELIMINARY TESTS FOR OPTIMUM MIX PROPORTION

To determine the mix proportions of sulfur concrete, the test results by Sheen et al. [**13**] “Preparation of modified sulfur concrete pipe using centrifugal force” were used. The research showed the most stable compressive and flexural strengths when the ratio of SPB and aggregate is 1:2 by volume. It is also presented that sulfur concrete had the best formability at 45% of the sand/aggregate ratio. Therefore, coarse and fine aggregates take 36.7 and 30 % respectively by volume if the total volume of sulfur concrete is considered as 100 %. The result shows that when 20 % of SPB was replaced with fly ash, the sulfur concrete has highest strength and proper workability. Therefore SPB and fly ash take 26.6 and 6.7 % respectively as a volume ratio. The weight amount of each component was calculated by multiplying specific gravity of each material.

I. Test Variables and Mixing Proportions

Based on the results of the aforesaid research, 7 cases of specimens as shown in Table 2 were tested with different types and sizes of aggregate, and various proportions of fly ash and SPB to find several optimal mix proportions. Three different maximum size of the coarse aggregate (13, 19, 25 mm) were used to ensure workability and strength. Amount of the fly ash was increased considering price of the sulfur concrete by minimizing the amount of SPB with preserving workability. Specimens of no.6 case were designed to ensure a possibility of using recycled aggregate as a coarse aggregate. The specimen no. 3 which has the same material proportion with specimen no. 4 was designed to confirm effect of curing age on mechanical strength.

Table 2: Test variables and mixing proportion

Specimen	Coarse aggregate (%)		Fine aggregate (%)		SPB (%)		Fly ash (%)		Maximum size of coarse aggregate (mm)	Type of coarse aggregate	Curing age (days)
	vol	wt	vol	wt	vol	wt	vol	wt			
NO.1	36.4	40.2	30.0	32.3	27.6	22.1	6.0	5.4	25	Natural	3
NO.2	36.4	40.2	30.0	32.3	27.6	22.1	6.0	5.4	13	Natural	3
NO.3	36.4	40.2	30.0	32.3	27.6	22.1	6.0	5.4	19	Natural	30
NO.4	36.4	40.2	30.0	32.3	27.6	22.1	6.0	5.4	19	Natural	3
NO.5	36.4	39.8	30.0	32.1	19.3	15.3	14.3	12.8	19	Natural	3
NO.6	36.4	35.0	30.0	34.6	19.3	15.4	14.3	15.0	25	Recycled	3
NO.7	36.4	39.7	30.0	31.9	15.2	12.0	18.4	16.4	19	Natural	3

II. Materials

In this study, DCPD-modified sulfur produced from Micro Powder was used to make sulfur concrete specimens. The property of SPB is presented in Table 3.

Table 3: Property of SPB

Element content (%)			Density (g/cm³)
S	C	O	
94~95	2.3	0.1	1.9

The properties of fly ash as a mineral filler of the preliminary test are shown in Table 4.

Table 4: Property of fly ash used in the preliminary tests

SiO2 (%)	Moisture (%)	Loss on ignition (%)	Specific gravity (g/cm³)	Specific area (cm²/g)
48.8	0.1	3.5	2.14	3360

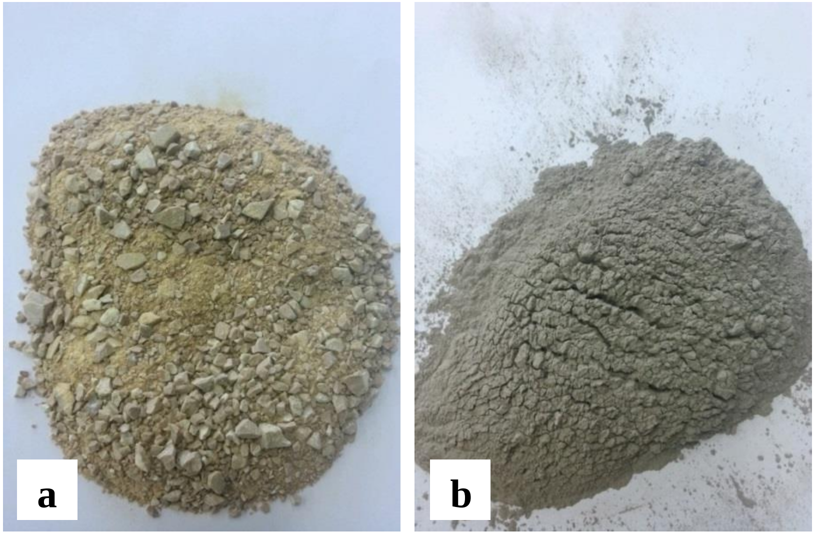


Figure 5: (a) SPB and (b) fly ash

The properties of coarse and fine aggregates are presented in Table 5. The recycled coarse aggregates used have a lower density and a higher water absorption ratio than the natural coarse aggregates. The particle size distributions of the used aggregates are presented in Figure 7. The natural coarse aggregates are crushed aggregates (Figure 6) that have the maximum size of 25, 19, and 13 mm. The maximum size of recycled coarse aggregates and natural fine aggregate are 25 and 10 mm respectively.

Table 5: Properties of aggregates used in the preliminary tests

Property of aggregate	Used aggregates			Test regulation
	Recycled coarse aggregate	Natural coarse aggregate	Fine Aggregate	
Absolute dry density (g/mm^3)	2.14	2.62	2.56	KS F 2503
Absorption (%)	6.28	0.84	1.41	KS F 2503
Abrasion (%)	21.1	14.6	-	KS F 2508
Absolute volume (%)	57	59	58	KS F 2527

0.08 mm sieve passing (%)		0.6	0.2	1.6	KS F 2511
Alkali aggregate reaction		harmless			KS F 2545
Amount of clay mass (%)		0.15	0.08	0.4	KS F 2512
Stability (%)		4.9	2.4	3.5	KS F 2507
Contents of impurity (%)	Organic impurity	Less than 1.0 (volume)	-	-	KS F 2576
	Inorganic impurity	Less than 1.0 (weight)	-	-	

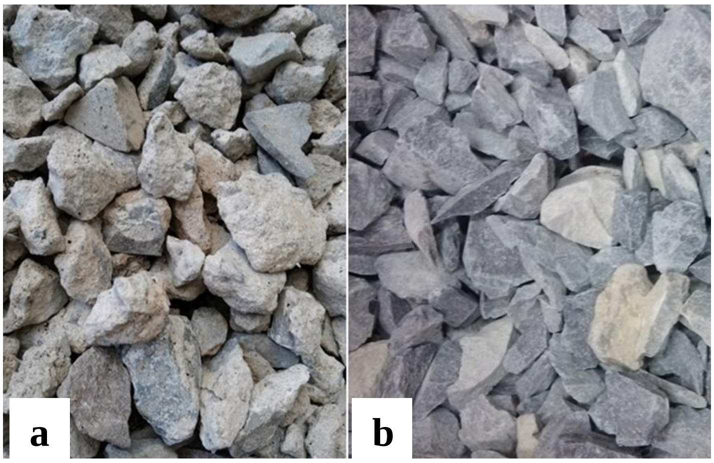


Figure 6: Coarse aggregates used in the tests (a) recycled, (b) natural

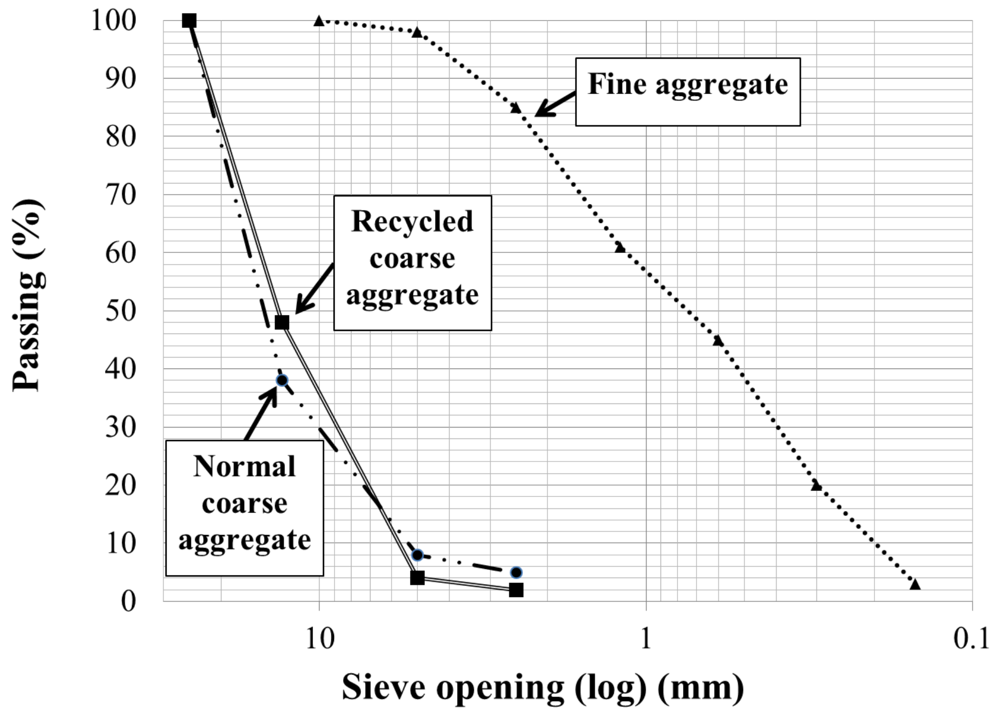


Figure 7: Particle size distributions of coarse and fine aggregates used in the preliminary tests

III. Result of optimum mixing proportions

For the specimens made with the maximum size of 19 mm, 13 mm, and 25 mm coarse aggregate, the average compressive strength was 76, 53, and 50 MPa, respectively. When the proportion of fly ash was increased to 5, 12, and 15% as a replacement of SPB, the compressive strength of sulfur concrete showed 76, 83, and 72 MPa, respectively. Therefore, the case with 19 mm coarse aggregate and 15% fly ash (by weight) presented the best mechanical properties preserving workability.

1-1. Compressive and splitting tensile strength

Table 6: Compressive and splitting tensile strength

Specimen	Density (kg/m^3)	Compressive strength (MPa)	Splitting tensile strength (MPa)
NO.1	2406	50	-
NO.2	2430	53	-

NO.3	2451	70	-
NO.4	2444	76	5.4
NO.5	2454	83	6.1
NO.6	2359	82	4.3
NO.7	2438	72	4.5

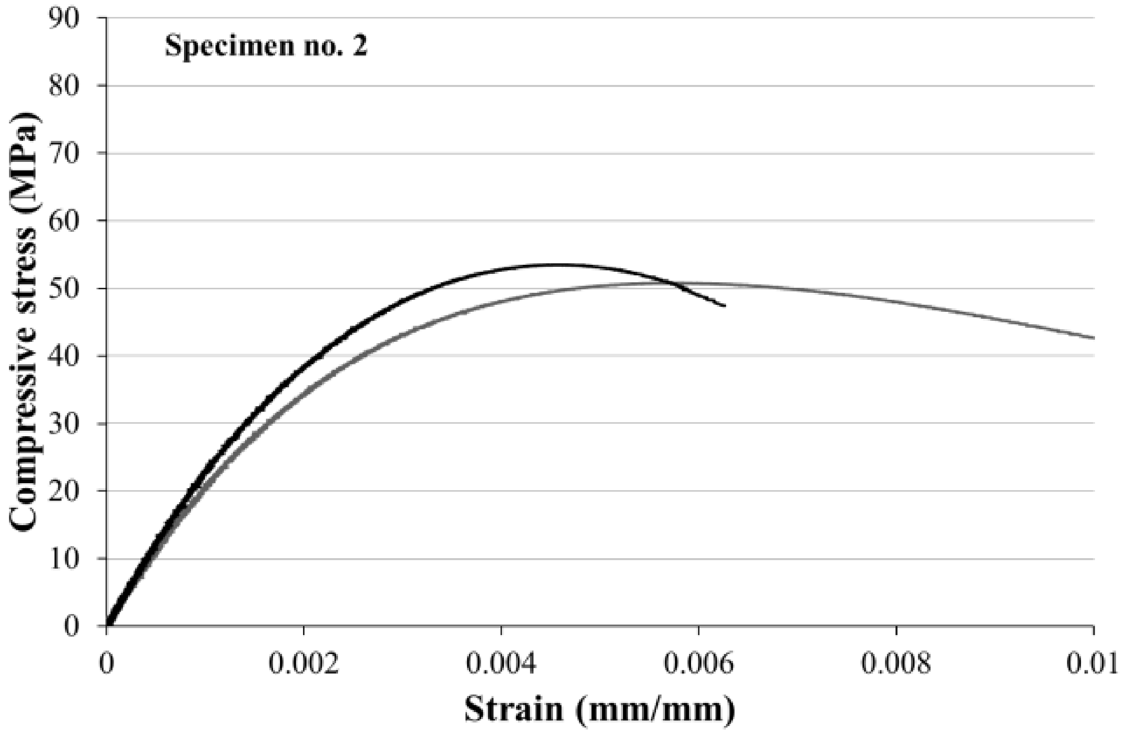
Specimen no. 1, 2, and 4 were compared each other to confirm effect of size of coarse aggregate on strength. The specimen no. 4 which is mixed with coarse aggregate of maximum size 19 mm showed the best performance of the strength among three specimens (Specimen no. 1, 2, and 4). The reason of reduced strength of specimen no. 1 is expected that large size of coarse aggregate caused bad workability. The possible reason of that specimen 2 had low strength might be improper particle size distribution of coarse aggregate.

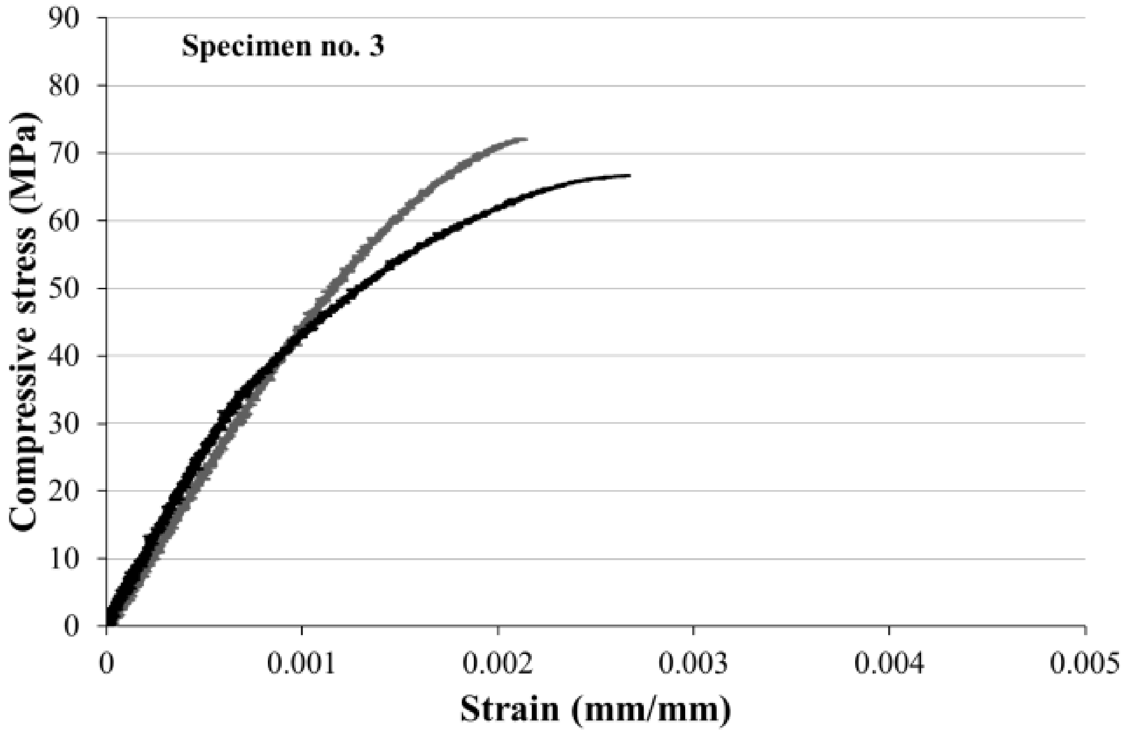
Compressive strengths of specimen no. 3 and 4 were compared to find effect of curing age of sulfur concrete. As a result, there was no difference of compressive strength between specimen no. 3 cured during 30 days and specimen no. 4 cured for 3 days. The little difference of 7 MPa of compressive strength between two specimens may be result of deviation of specimens. This means that sulfur concrete reach to final strength in early age and does not need to long curing time as Portland cement concrete.

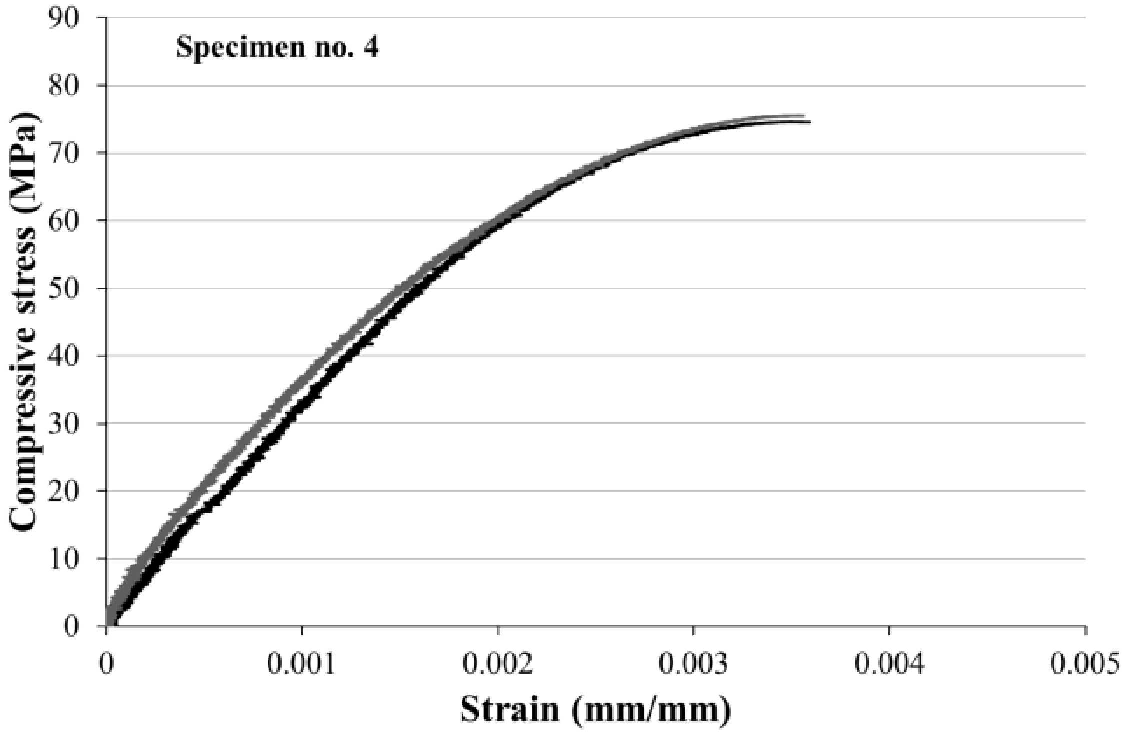
In order to determine proper proportion of fly ash and SPB, the strengths of specimen no. 4, 5, and 7 were measured and compared to each other. Specimen no. 5 which is mixed with 15 % SPB and 12 % fly ash obtained the highest compressive and splitting tensile strength among all specimens. The test result shows that an increase in the proportion of fly ash while reducing amount of SPB is helpful to improve the strength of the sulfur concrete. These results are attributed to well particle size distribution improved by increasing fly ash. However, strength was decreased when amount of SPB was reduced to 12 % because amount of SPB as a plastic state decide to the workability.

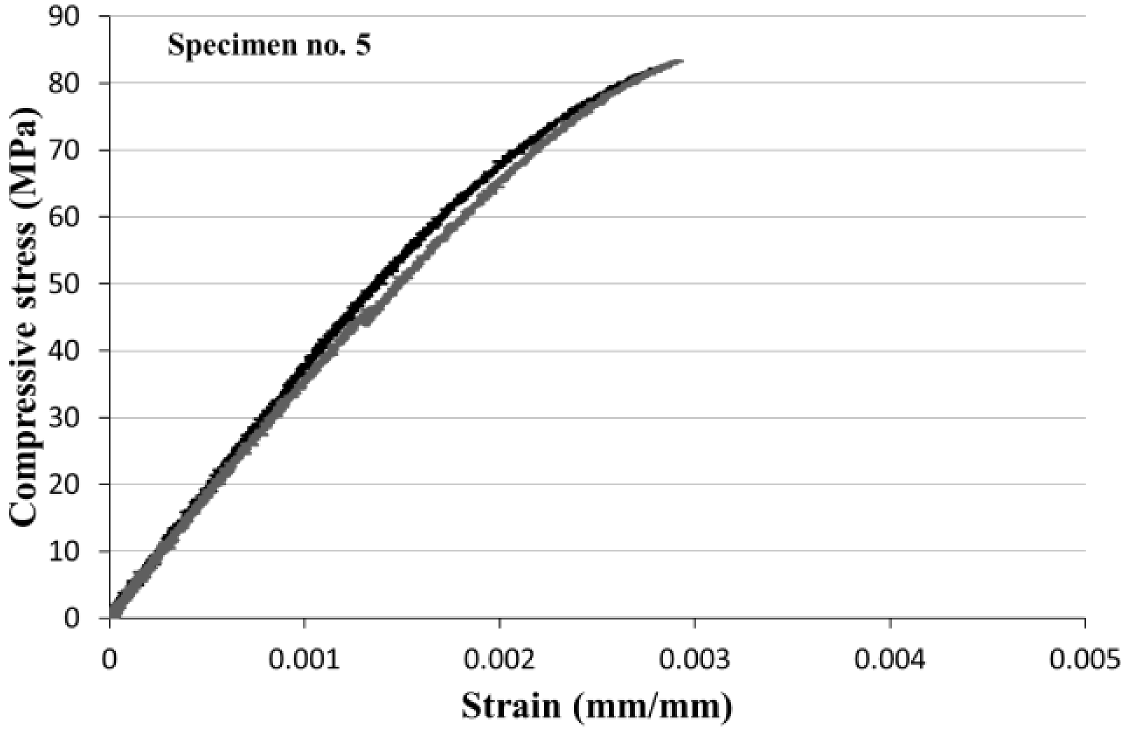
The specimen no. 6 was fabricated using recycled coarse aggregate of maximum size 25 mm to examine an applicability of recycled aggregate to sulfur concrete. The splitting tensile strength of the specimen no. 6 was decreased about 30 % when it compared with specimen no. 5. However, there was no difference in compressive strength between two specimens.

1-2. Elastic modulus


Specimen no. 2
Compressive stress (MPa)
Strain (mm/mm)
0
10
20
30
40
50
60
70
80
90
0
0.002
0.004
0.006
0.008
0.01


Specimen no. 3
Compressive stress (MPa)
Strain (mm/mm)
0
10
20
30
40
50
60
70
80
90
0
0.001
0.002
0.003
0.004
0.005


Specimen no. 4
Compressive stress (MPa)
Strain (mm/mm)
0
10
20
30
40
50
60
70
80
90
0
0.001
0.002
0.003
0.004
0.005


Specimen no. 5
Compressive stress (MPa)
Strain (mm/mm)
0
10
20
30
40
50
60
70
80
90
0
0.001
0.002
0.003
0.004
0.005

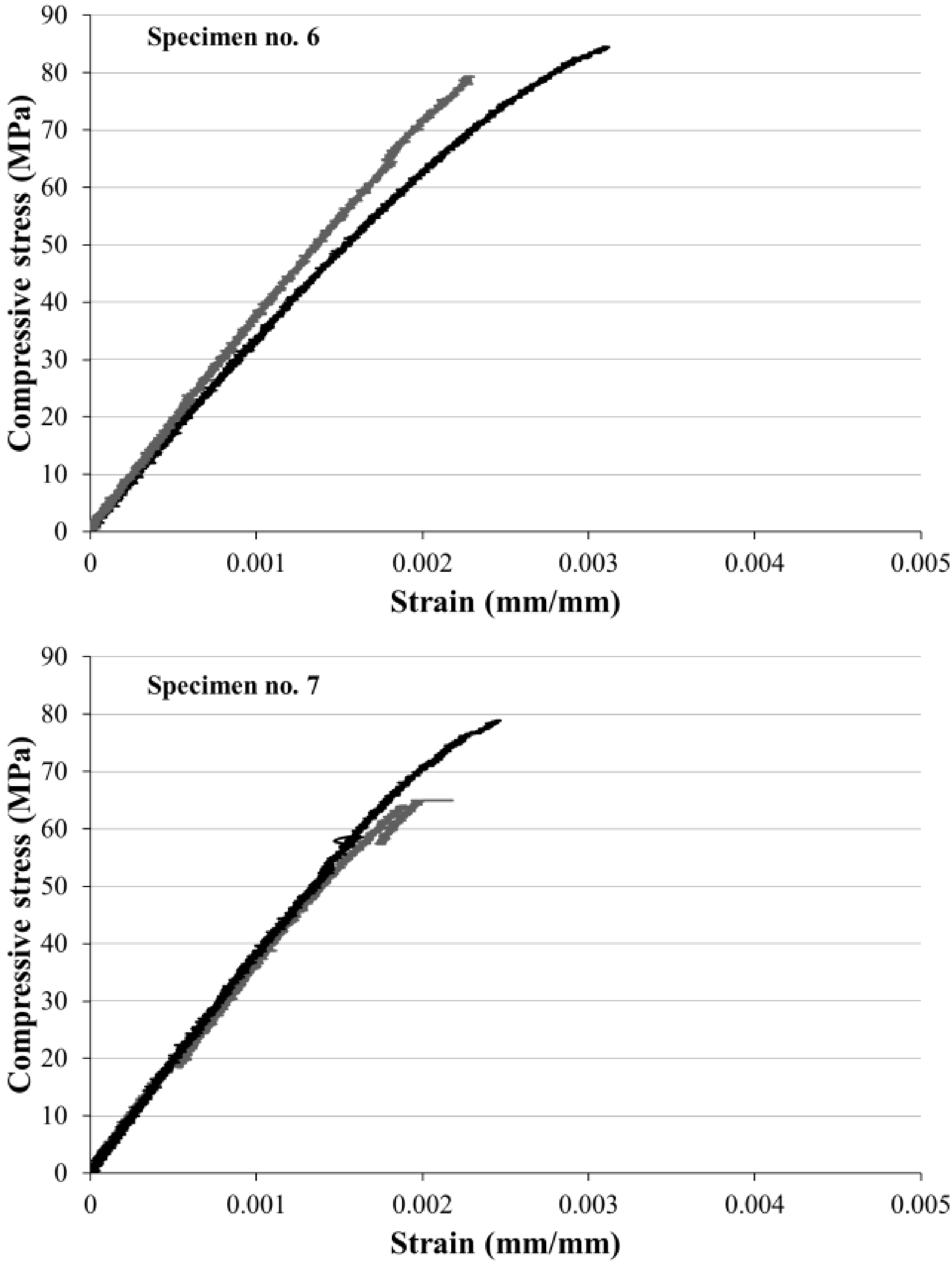


Figure 8: Stress- strain curves

Table 7: Elastic modulus

Specimen	Strain at maximum stress	E_{exp} (GPa)	E_{code} (GPa)	E_{exp}/E_{code} (%)
NO.1	-	-	-	-
NO.2	0.0052	21.3	34.2	62
NO.3	0.0024	48.7	39.4	124
NO.4	0.0035	35.4	41.0	86
NO.5	0.0031	36.4	43.1	84
NO.6	0.0027	35.6	42.8	83
NO.7	0.0024	37.3	40.1	93

* E_{exp} is measured at 40% of $f_{c\ max}$

* $E_{code} = 4730 \sqrt{f_{c\ max}}$ [14]

The stress strain curves of specimens are shown in Figure 8. Elastic modulus and strain at maximum stress are presented in Table 7. The average strain of sulfur concrete at maximum stress is 0.0032. The elastic modulus of sulfur concrete was compared with calculated elastic modulus of Portland cement concrete. The average elastic modulus of sulfur concrete is 89 % of that of ordinary Portland cement concrete.

CHAPTER 5. TESTS FOR MACHANICAL AND DURABILITY PROPERTIES

I. Test Variables and Mix Proportions

Based on the results of the preliminary tests, three types of specimens as shown in Table 8 were fabricated in different mix proportions.

Table 8: Test variables and mix proportions

Specimen type	Coarse aggregate (%)		Fine aggregate (%)		SPB (%)		Fly ash (%)		Maximum size of coarse aggregate (mm)	Type of coarse aggregate
	vol	wt	vol	wt	vol	wt	vol	wt		
S	36.4	40.5	30.0	32.5	33.6	26.9	0.0	0.0	19	Natural
F	36.4	39.6	30.0	31.8	19.1	15.0	14.5	13.6	19	Natural
R	36.4	34.6	30.0	34.3	17.7	15.0	15.9	16.1	19	Recycled

The effect of using fly ash on mechanical and durability properties is investigated by comparing S-type and F-type specimens. F-type specimens were made with a reduced amount of SPB and 14% of fly ash with maintaining the proportion of aggregate. R-type specimens were prepared to examine the applicability of recycled aggregate for sulfur concrete.

II. Materials

In these tests, the same type of modified sulfur with that used in the preliminary tests was used as a binder. The property of SPB is presented in Table 3.

The properties of fly ash used as a mineral filler in these tests are shown in Table 9. A different type of fly ash was used from the preliminary tests.

Table 9: Property of fly ash

SiO2 (%)	Moisture (%)	Loss on ignition (%)	Specific gravity (g/cm^3)	Specific area (cm^2/g)
52.4	0.34	2.2	2.28	3862

The properties of coarse and fine aggregates are presented in Table 10. The recycled coarse aggregates used have the same properties with those used in the primary tests as shown in Table 5. The particle size distributions of the used aggregates are presented in Figure 9. The maximum size of both natural and recycled coarse aggregates is 19 mm. The natural coarse aggregate is crushed granite and maximum size of natural fine aggregate are is 10 mm.

Table 10: Properties of aggregates

Property of aggregate	Used aggregates			Test regulation
	Recycled coarse aggregate	Natural coarse aggregate	Fine Aggregate	
Absolute dry density (g/mm^3)	2.14	2.64	2.57	KS F 2503
Absorption (%)	6.28	0.86	1.36	KS F 2503
Abrasion (%)	21.1	14.4	-	KS F 2508
Absolute volume (%)	57	59	57	KS F 2527
0.08 mm sieve passing (%)	0.6	0.4	1.8	KS F 2511
Amount of clay mass (%)	0.15	0.09	-	KS F 2512
Stability (%)	4.9	2.8	3.4	KS F 2507

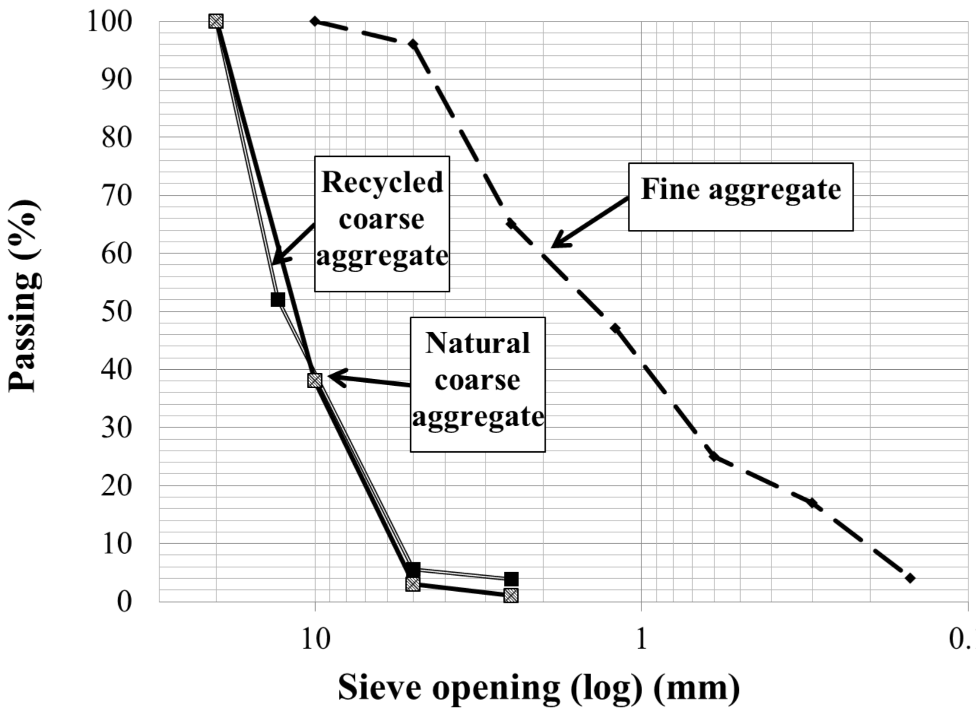


Figure 9: Particle size distributions of coarse and fine aggregates

III. Test results

Tests for compressive and splitting tensile strengths, freezing and thawing resistance, thermal expansion, and chemical resistance were performed. The test results are presented in the following.

1-1. Compressive and splitting tensile strength

Table 11: Compressive and splitting tensile strengths

Specimen	Density (kg/m^3)	Compressive strength (MPa)		Splitting tensile strength (MPa)	
			Average		Average
S	2389	62.4	63.3	3.3	3.6
		64.1		3.9	
F	2447	90.6	88.0	6.6	6.6
		85.3		6.5	
R	2302	75.5	77.6	4.4	4.4
		77.7		4.3	

Two specimens which were cured for 93 days were tested for each of the compressive and tensile strengths. The average strength of two specimens each case was presented in Table 11. The F-case specimens presented the highest compressive and splitting tensile strengths. As a result of using fly ash and reducing the amount of SPB, the compressive strength increased by39.7 % and the tensile strength increased by 83.3%. ; compare specimens S and F. It is expected that fly ash influenced to increase the density of the sulfur concrete by filling the pores. The recycled coarse aggregate caused 12.5 and 33.3 % reductions in compressive and tensile strengths respectively, compared with specimen F. A possible reason for the strength reduction caused by recycled coarse aggregate is that attached mortar and cement paste around the coarse aggregate decreased the density of the concrete. On the other hand, the specimen R was higher than that of specimen S. However, the compressive strength of S-case which is measured before immersion of acid and salt environments is slightly higher than that of R-cases. This tendency of the results is probably due to the deviation of the quality of sulfur concrete.

1-2. Elastic modulus

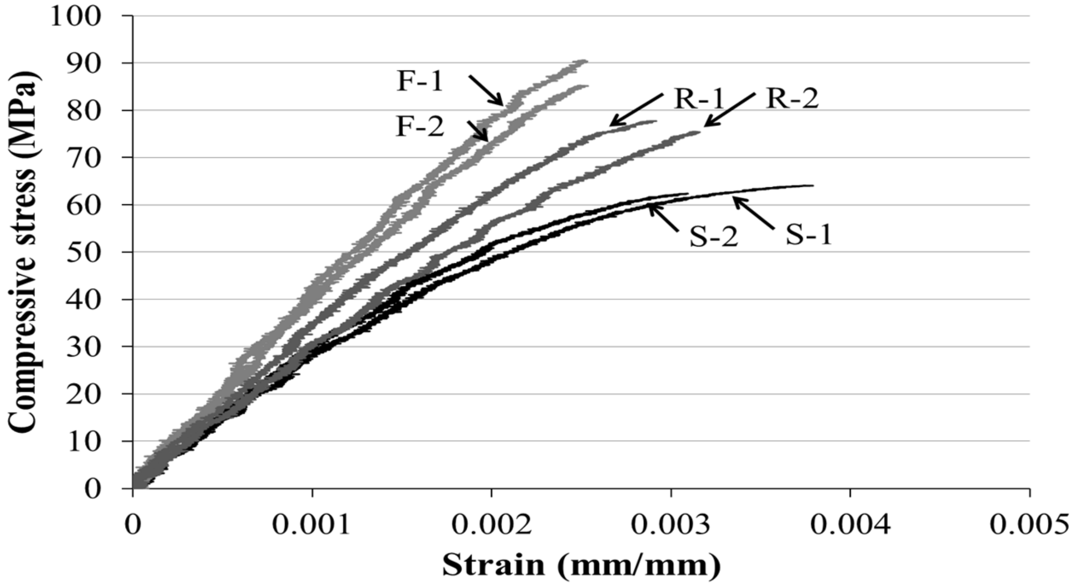


Figure 10: Stress-strain curves

Table 12: Elastic modulus of sulfur concrete

Specimen	Strain at max stress	E_{exp} (GPa)	E_{code} (GPa)	$E_{exp/}$ E_{code} (%)
S	0.0334	29.1	37.6	77
F	0.0025	40.4	44.4	81
R	0.0030	32.5	41.4	78

* E_{exp} is measured at 40% of $f_{c\ max}$

* $E_{code} = 4730\ \sqrt{f_{c\ max}}$ [14]

The stress-strain curves of sulfur concrete with different mixing proportions are shown in Figure 10. In Figure 10, it can be confirmed that the maximum compressive strain decreases with the increase of compressive strength. Thus, F-case specimens and S-case specimens presented the lowest and highest strain under the maximum stress respectively.

The strain of R-case specimens was higher than that of F-case specimens under same loads. This is mainly due to presence of interface between old cement mortar-aggregate and old cement mortar-SPB. Those interfaces in concrete may give rise to a progressive development of micro-cracks. Therefore, the strain of the concrete containing recycled aggregate increases at a faster rate than the concrete made with natural aggregate. [**15**]

Elastic modulus of three cases of sulfur concrete is presented in Table 12. E_{exp} is the secant elastic modulus which is calculated from the experiment and E_{code} is calculated elastic modulus using ACI318-11 [14] based on compressive strength of ordinary Portland cement concrete. The results shows that elastic modulus of sulfur concrete is generally lower than that of ordinary Portland cement concrete which have the same compressive strength with sulfur concrete. This means that sulfur concrete have bigger strain than ordinary Portland cement concrete under the same stress.

1-3. Chemical resistance

In order to compare the strength change between Portland cement concrete and sulfur concrete, the test results of Portland cement concrete from earlier two papers are presented in Figure 11. The dashed and solid lines stand for the results from Yoon [**16**] and Vlahovic et al. [**19**] respectively. The specimens of Yoon [**16**] were made with the maximum size of coarse aggregate equal to 25 mm and the water/cement ratio of 35%. The specimens of Vlahovic et al. [**19**] were made with only fine aggregates of which the maximum size is 2 mm and with the water/cement ratio of 54%. The average compressive strength of the specimen by Yoon [**16**] is reduced by 50% after immersion in three different solutions for 30 days compared with that after 7 days. The result of test performed by Vlahovic et al. [**19**] show that the average compressive strength was decreased by 95% after immersion of 30 days. The mechanism of chemical reaction between Portland cement concrete and acid solution is presented below.

Sulfuric acid creates gypsum by reacting with calcium hydroxide which is generated from cement hydration. The gypsum reacts with calcium aluminate to create ettringite. The ettringite causes expansion pressure which lead to cracking and collapse of concrete.

Ca(OH)2 + H2SO4 → CaSO4 • 2H2O + H2O

CaSO4 • 2H2O + 3CaO • Al2O3 • 6H2O → 3CaO • Al2O3 • 3CaSO4 • 31~32H2O

For hydrochloric acid case, mechanism of reaction with cement hydrate is shown below.

Ca(OH)2 + HCl → CaCl2 • 2H2O + H2O

3CaO • 2SiO2 • 3H2O + HCl → CaCl2 • SiO2 • H2O

3CaO • Al2O3 • CaSO4 • 32H2O + HCl → CaCl2 + Al2O3 • nH2O + CaSO4 • 2 H2O

The cement hydrate loses its combining ability by above reaction. CaCl2 which is well-soluble is easily dissolved. SiO2 and Al2O3 also are dissolved as a gel state.

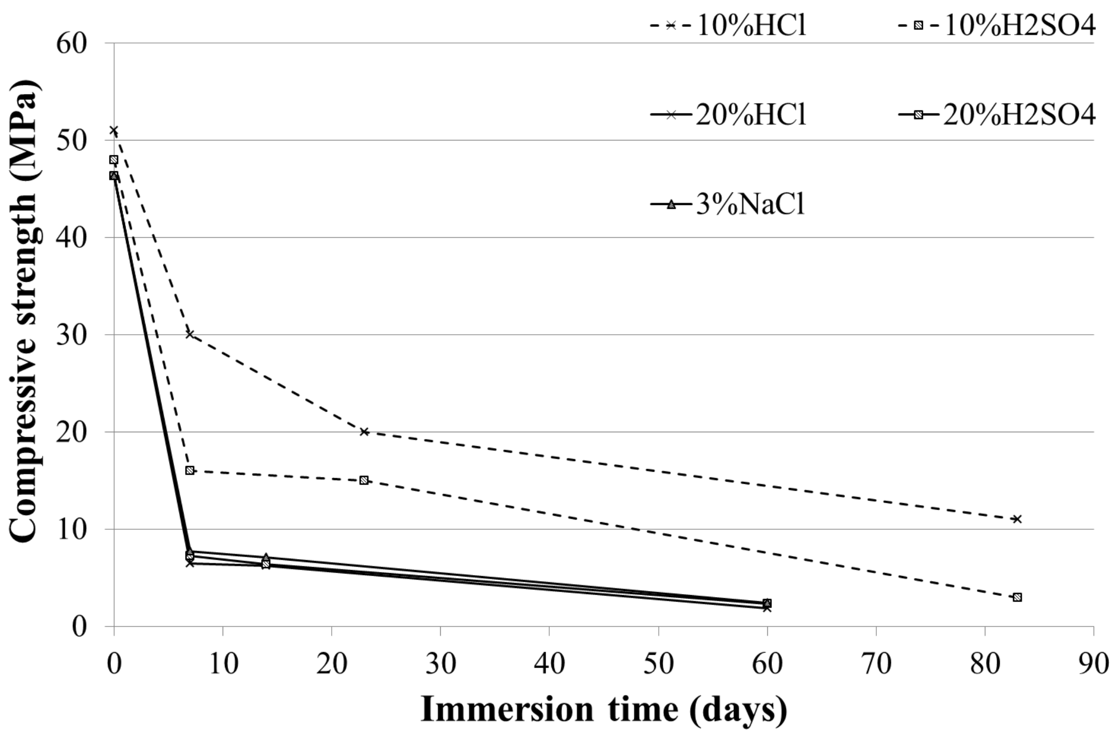


Figure 11: strength change of Portland cement concrete in acid and salt solutions [16, 19]

The result of compressive strength of sulfur concrete after immersion in acid and salt solution is shown in **Table 13** and Figure 12. In this test, the average compressive strength of sulfur concrete was decreased 10 % after immersion of 60 days. The specimens tested by Vlahovic et al. [**19**] are made with only fine aggregate which maximum size is 2 mm having 30 % of SPB and 7 % of fly ash. The result shows that compressive strength of these specimens was decreased up to 2~3% by HCl and H_2SO_4 solutions and NaCl solution did not reduce the strength of sulfur concrete after immersion of 360 days. The reason of strength reduction in this experiment is expected that small amount of SPB and use of coarse aggregate influenced the resistance of acid and salt solution.

Table 13: Change of compressive strength after 60 day of immersion in different solutions

	Change of compressive strength after 60 day of immersion in different solutions						
	Before (MPa)	10% HCl		20% H2SO4		3% NaCl	
		After (MPa)	Reduction ratio (%)	After (MPa)	Reduction ratio (%)	After (MPa)	Reduction ratio (%)
F-1	80.8	67.0	17.1	67.1	17.0	65.6	18.8
F-2	76.8	68.8	10.4	64.6	15.9	62.1	19.1
F-3	77.3	62.6	19.1	66.0	14.7	62.0	19.9
R-1	61.1	60.0	1.8	62.9	0.0	62.7	0.0
R-2	60.5	61.2	0.0	60.1	0.6	61.1	0.0
R-3	62.7	59.5	5.1	60.3	3.9	60.6	3.4
S-1	65.9	59.1	10.3	54.9	16.8	55.0	16.6
S-2	62.2	58.1	6.6	55.0	11.5	53.2	14.4
S-3	63.7	60.7	4.8	54.2	14.9	54.1	15.1

Table 14: Test results from similar experiment

Test results by Vlahovic et al. [19]	Days of immersion in different solutions (days)						
	0	7	14	21	110	220	360
	Compressive strength (MPa)						
10% HCl	48.9	48.4	48.2	48	47.9	47.7	47.4
20% H_2SO_4	48.9	48.5	48.4	48.1	47.8	47.9	47.9
3% NaCl	48.9	48.7	48.9	49.2	48.9	49.0	49.0

Test results by Yoon [16]	Days of immersion in different solutions (days)				
	0	7	23	83	173
	Compressive strength (MPa)				
10% HCl	54	43	57	59	48
10% H_2SO_4	62	59	60	40	48
10% NaCl	50	48	64	50	51

In this experiment, the F-case showed the most significant strength reductions as 17 %, while the R case presented the smallest strength reductions as 2 % in the three different solutions. Strength of F case was decreased up to 19 % by NaCl solution. However, strength of R case was decreased up to 2 % by HCl solution. The average strength reduction of S-case is 12 % in all solutions. More research will be needed to find reasons of small strength reduction of R-case.

The mass change of sulfur concrete is shown in Figure 13. The F-case and R-case showed 0.4 % of average mass reductions, while the S-case presented only 0.1 % of mass reductions in the three different solutions. Mass of R-case was decreased up to 1.1 % by HCl solution and mass of R-case was decreased up to 0.1 % by H_2SO_4 solution. In NaCl solution, mass of F-case and R-case specimens are slightly increased up to 0.1 % and 0.2 % respectively. However, there was no mass change of S-case in Nacl solution. The mechanism of reaction with sulfur concrete in acid environment mentioned by Vlahovic et al. [**19**] is stated below.

It is well known that hydrochloric and sulfuric acids do not affect sulfur. The aggregate and fillers used in the manufacture of sulfur concrete are constituted by mineral oxides. The attack of sulfur concrete by hydrochloric or sulfuric acid solution is based on the reactions of basic and amphoteric oxides with acids and resulted in the formation of metal chlorides or sulfates. Most bases dissolve in water, and thereby release hydroxide ions (OH-). Hydroxide ions react with acids and form salts. Hydrogen ions are accepted by a calcium oxide base; hence a base is a proton acceptor.

Acid + Basic Oxide → Salt + Water

H_2SO_4 (aq) + CaO (s) → $CaSO_4$ (aq) + H_2O (l)

Amphoteric aluminum oxide reacts in the same way:

Acid + Amphoteric Oxide → Salt + Water

6HCl (aq) + Al_2O_3 (s) → $2AlCl_3$ (aq) + $3H_2O$ (l)

Sodium chloride does not react with sulfur or with the oxides present in the aggregate and fillers. Hence, physical processes on the contact regions sulfur/ aggregate and filler are responsible for the attack in the saline media. The growth of sodium chloride crystals leads to a partial detaching between sulfur and aggregate and filler.

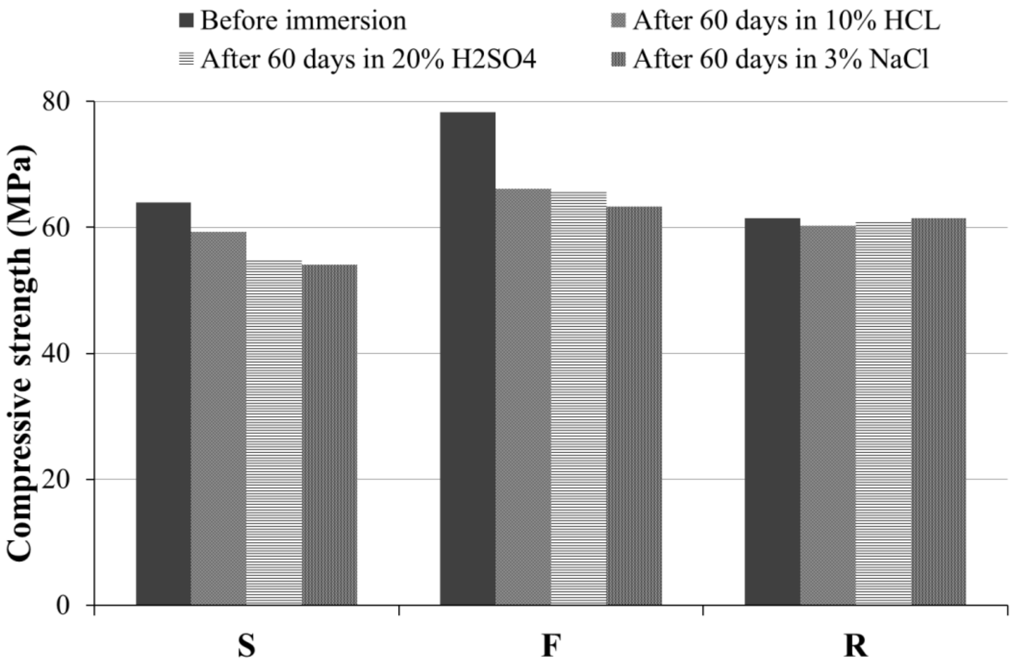


Figure 12: Compressive strength change by aggressive chemical immersion

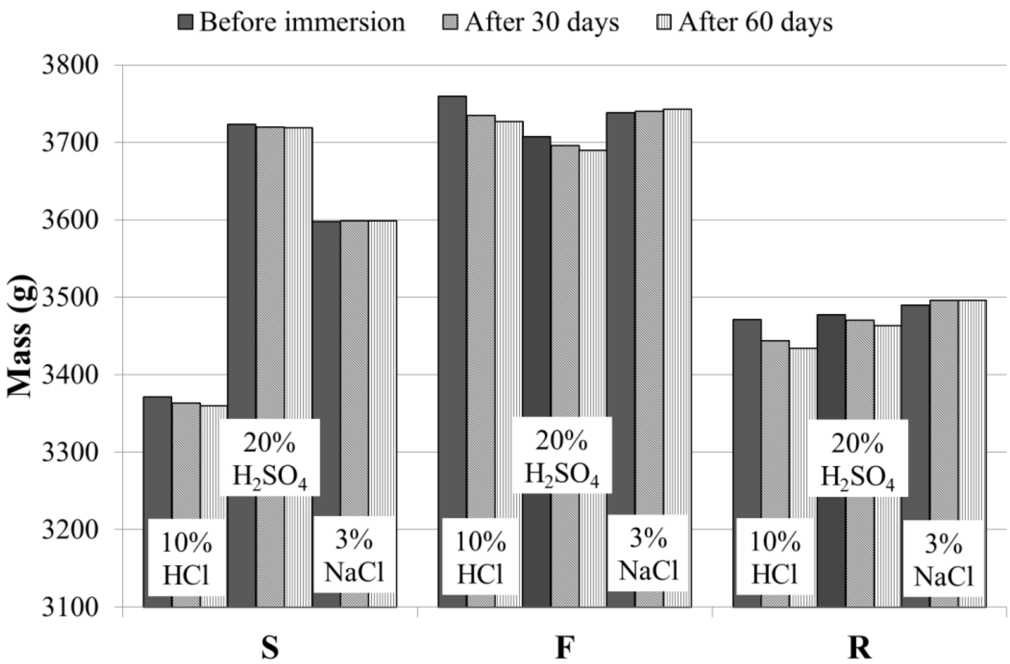


Figure 13: Mass change by aggressive chemical immersion

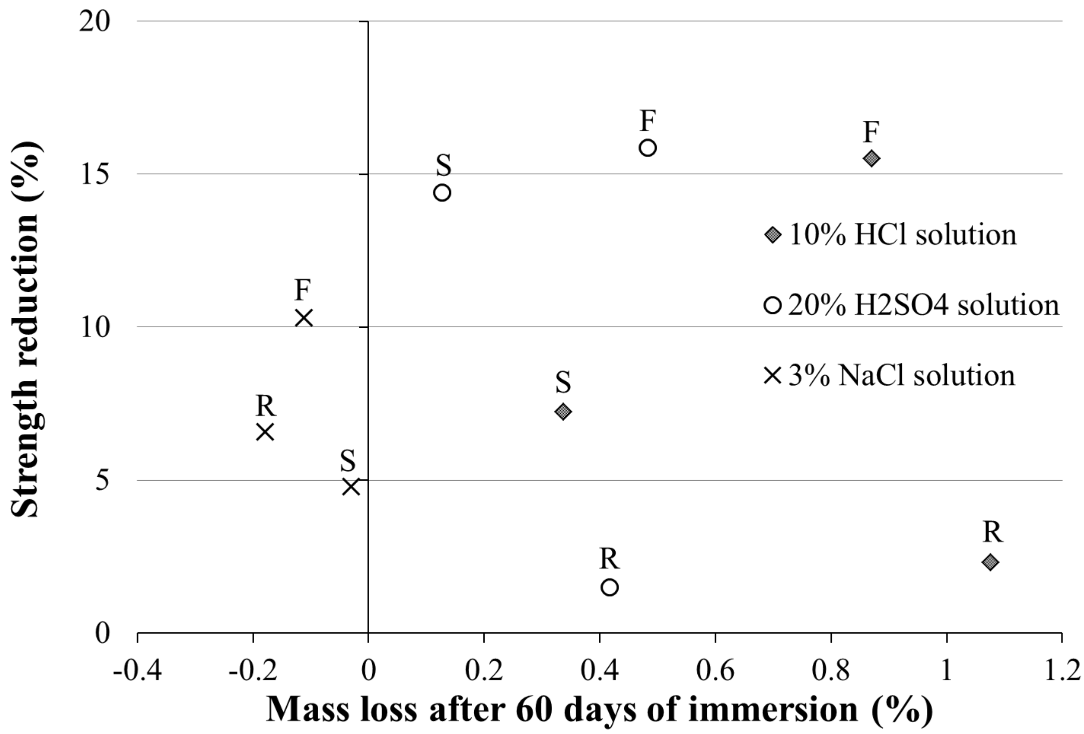


Figure 14: Rate of mass and strength change by aggressive chemical immersion

1-4. Coefficient of thermal expansion

The coefficient of thermal expansion of each case was presented in Table 15 and Figure 15. The average of the measured coefficients of thermal expansion of sulfur concretes is 15.26×10^{-6}/°C. This value is bigger than that of Portland cement concrete which has range from 10.0×10^{-6}/°C to 13.0×10^{-6}/°C. The influence of the mix proportions arises from the fact that the two main constituents of concrete, hydrated cement paste and aggregate, have dissimilar thermal coefficients, and the coefficient of concrete is a resultant of the two values. [17]

The F-case specimen showed most similar value with the coefficient of normal concrete. The 17 % of coefficient of thermal expansion of sulfur concrete was reduced by mixing fly ash when F-case specimens are compared with S-case specimens. It would be explained that SPB has higher coefficient of thermal expansion than that of fly ash. Elemental sulfur has a relatively high linear coefficient of thermal expansion 74×10^{-6}/°C. [5] Meyer also found that thermal expansion of fibrous sulfur is 94×10^{-6}cm/°C for the *a* axis and 72×10^{-6}cm/°C for the *b* axis. [18] Using recycled coarse aggregate cause slight increase of coefficient of thermal expansion when the R-case and F-case were compared. Typical values of the coefficient of cement paste vary between $11{\sim}20\times10^{-6}/^{\circ}C$. Therefore, it is expected that attached cement paste and mortar around recycled aggregate influenced increase of coefficient of thermal expansion of R-case. Kim et al. [**21**] also mentioned that recycled aggregate caused increase of coefficient of thermal expansion because of effect of attached mortar around the aggregate.

Table 15: Coefficient of thermal expansion of each case

Coefficient of thermal expansion	
S	$17.2\times10^{-6}/^{\circ}C$
F	$13.8\times10^{-6}/^{\circ}C$
R	$14.8\times10^{-6}/^{\circ}C$

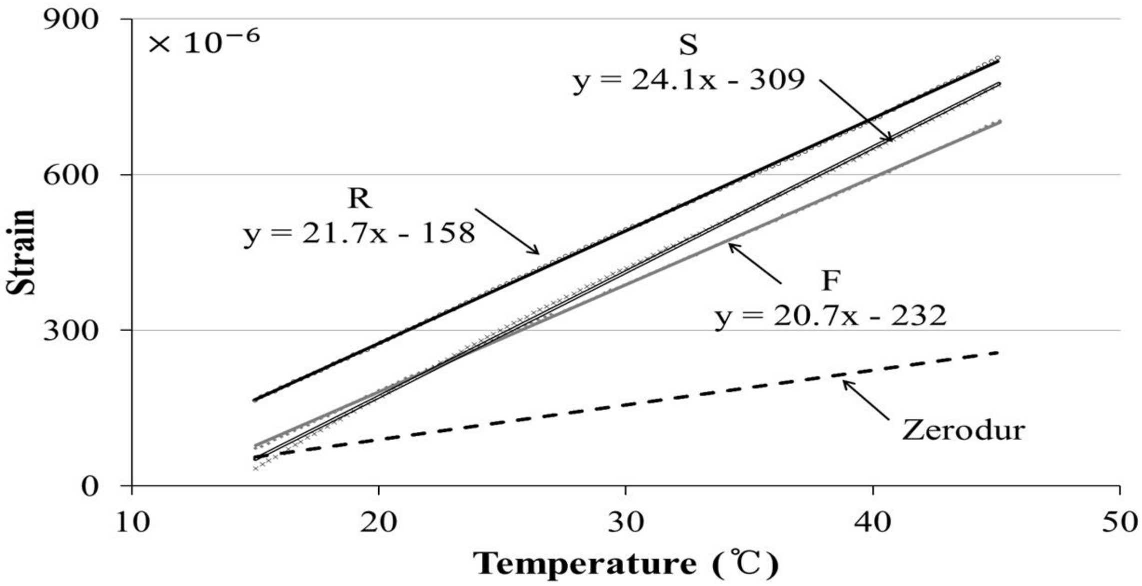


Figure 15: Measured thermal expansion

1-5. Resistane of freezing and thawing

Table 16: Relative dynamic modulus of elasticity

Specimen	Relative dynamic modulus of elasticity (%)					
	After 100 cycles	Average	After 200 cycles	Average	After 300 cycles	Average
F-1	94.3	93.1	88.7	87.6	86.5	85.0
F-2	92.5		85.6		83.1	
F-3	92.7		88.4		85.4	
R-1	90.2	89.5	83.7	82.6	75.4	77.6
R-2	89.1		81.1		76.7	
R-3	89.2		83.0		80.6	
S-1	92.7	91.7	86.2	87.2	83.4	84.1
S-2	91.7		86.7		83.2	
S-3	90.7		88.8		85.7	

Sulfur concrete resistance to frost action is of importance for structures that are subjected to such climatic condition. Under moist condition, it is important that the material endure frost action. The measured relative dynamic modulus of elasticity of three cases is shown Figure 16 and Table 16. The tested sulfur concretes presented high resistance to freezing and thawing. S- and F-case specimens made with natural aggregates had 84.6 % of relative dynamic elastic modulus after 300 cycles of temperature change. However, R-case specimens made with recycled aggregates showed 77.6 %.

According to Mehta and Monteiro [11], it is obvious that the ability of a normal concrete to resist damage due to frost action depends on the characteristics of both the cement paste and the aggregate. In each case, the outcome is controlled actually by the interaction of several factors, such as location of escape boundaries (distance by which water has to travel for pressure relief), the pore structure of the system (size, number, and continuity of pore), the degree of saturation (amount of freezable present), rate of cooling, and the tensile strength of the material that must be exceeded to cause rupture. The provision of escape boundaries in cement paste matrix and modification of its pore structures are the two parameters that are relatively easy to control. The provision of escape boundaries can be controlled by means of air entrainment in concrete and pore structures can be modified by the use of proper mix proportions and curing.

McBee et al. [9] indicated that when the moisture absorption of sulfur concrete exceeds the limit (0.05 %), its resistance to freeze-thaw damage decreases dramatically. Therefore, good quality control for water absorption is essential in producing durable sulfur concrete. The key is a sufficient amount of SPB and a restricted amount of filler to attain the needed material density with a dense graded composition. Yoon [16] also tested resistance of freezing and thawing for 8 specimens of sulfur concrete specimens. He found that there was no mass change after 300 cycles and all specimens showed over 80 % of relative dynamic modulus of elasticity. He expected that the possible reason of high resistance of freezing and thawing cycles is low water absorption of sulfur concrete.

Measurement of a decrease in the mass of the specimen is appropriate when damage takes place mainly at the surface of specimen, but it is not reliable in case of internal failure; the result depend also in the size of the specimen. If failure is primarily due to unsound aggregate, it is more rapid and more severe than when the hardened cement paste is disrupted first.

Cohen [20] also tested freezing and thawing resistance of sulfur concrete using elemental sulfur binder. Cohen mentioned that analyses of the relative compressive strengths, surface conditions, and weight loss data of the water and air and water stored sulfur concrete seems to indicate good freeze and thaw resistance.

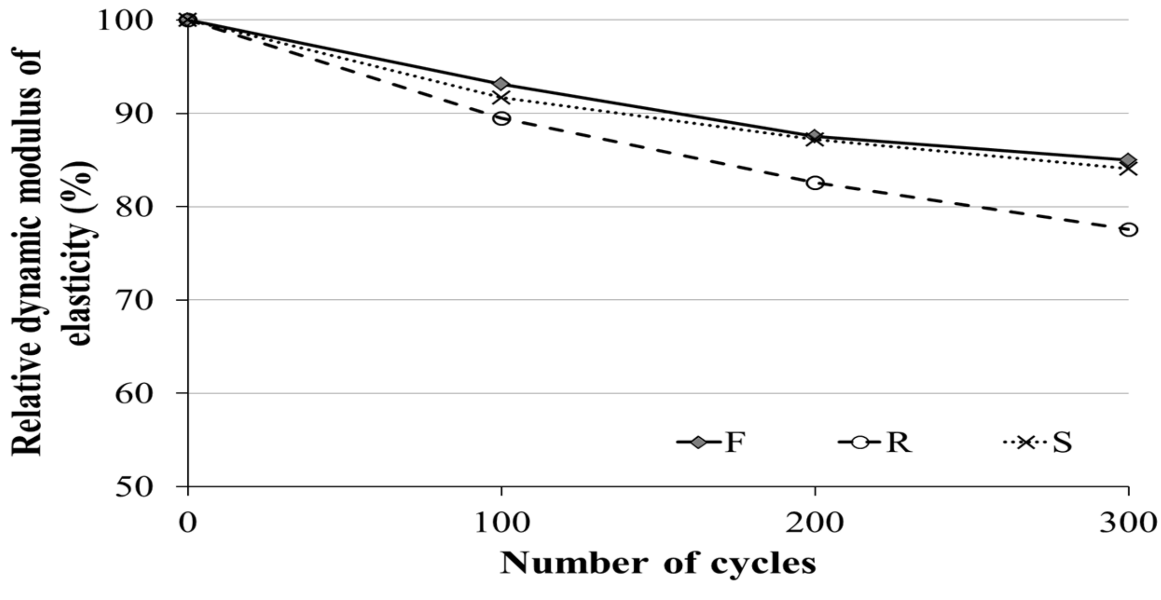


Figure 16: Resistance to Freezing and Thawing

In order to evaluate applicability of sulfur concrete to severe environment, concrete durability evaluation for freezing and thawing resistance suggested by KCI [**22**] was calculated. Durability evaluation for freezing and thawing resistance of concrete structures can be performed following below equations.

$$\gamma_P F_d \leqq \phi_K F_{lim}$$

γ_P: Environmental coefficient for freezing and thawing (usually 1.0)

$$F_d = \frac{1}{E_d}$$

$$F_{lim} = \frac{1}{E_{min}}$$

E_d: Relative coefficient of dynamic modulus of elasticity (%)

E_{lim}: Minimum relative coefficient of dynamic modulus of elasticity to satisfy freezing and thawing resistance of structure

ϕ_K: Coefficient of durability reduction for freezing and thawing by location and type of structures

Table 17: Minimum relative coefficient of dynamic modulus of elasticity [22]

Weather condition		Severe and frequent freezing and thawing		Moderate and infrequent freezing and thawing	
Thickness of cross section		Thin(2)	Normal	thin	Normal
Exposure state of structure	Continues and frequent saturation of water(1)	85	70	85	60
	Normal exposure (Not included above state)	70	60	70	60

(1) Saturated structures by water or water vapor

(2) Thickness of structure is less than 0.2m

Table 18: Coefficient of durability reduction for freezing and thawing [22]

	Normal part of structure	Upper part of structure
Normal structures	1.0	0.8
Important structures	0.9	0.7

Table 19: $\phi_K F_{lim}$

$\phi_K F_{lim}$	0.7	0.8	0.9	1
85	0.008	0.009	0.011	0.012
70	0.010	0.011	0.013	0.014
60	0.012	0.013	0.015	0.017

Table 20: $\gamma_P F_d$

Type of specimens	$\gamma_P F_d$
S	0.012
F	0.012
R	0.013

In this evaluation, environment is divided 12 cases. As a result of above evaluation, S- and F-case can be applied to 7 cases and R-case specimen can be used in 5 cases.

CONCLUSION

In this research, 7 cases of sulfur concrete were fabricated and tested with different types (natural and recycled) and sizes (13, 19, 25 mm) of aggregate, and various proportions of fly ash and SPB to find several optimal mix proportions as a preliminary test. Amount of the fly ash and SPB was adjusted to maintain proper price of the sulfur concrete by minimizing the amount of SPB with preserving workability. In order to secure the workability, unconsolidated sulfur concrete before casting was recorded by video camera.

As a result of confirmation, when smaller the maximum size of the coarse aggregate was used, better workability was found. There was no significant decrease of workability when the substitution ratio of fly ash increased up to 12 %. However, noticeable decrease of workability was found when the SPB was replaced with fly ash up to 15 %.

For the specimens made with the maximum size of 19 mm, 13 mm, and 25 mm coarse aggregate, the average compressive strength was 76, 53, and 50 MPa, respectively. When the proportion of fly ash was increased to 5, 12, and 15% as a replacement of SPB, the compressive strength of sulfur concrete showed 76, 83, and 72 MPa, respectively. Therefore, the case with 19 mm coarse aggregate and 15% fly ash (by weight) presented the best mechanical properties. The average strain of sulfur concrete at maximum stress is 0.0032. The elastic modulus of sulfur concrete was compared with calculated elastic modulus of Portland cement concrete. The average elastic modulus of sulfur concrete is 89 % of that of ordinary Portland cement concrete.

Based on the results of the preliminary tests, three types of specimens were fabricated in different mixing proportions. In order to investigate the effect of mixing with fly ash, S-case and F-case were compared. F-case specimens were mixed with reduced amount of SPB and 14% of fly ash maintaining proportion of aggregate. R-case specimens were fabricated to ensure applicability of recycled aggregate for sulfur concrete by comparing with F-case specimens.

As a result of strength test, the F-case specimens presented the highest compressive and splitting tensile strength. It is expected that fly ash influenced to increase the density of the sulfur concrete by improving the aggregate size distribution and filling the pores. The recycled coarse aggregate caused strength reduction at both compressive and tensile strength test compared with F-case specimens. A possible reason for strength reduction is that attached mortar and cement paste around the coarse aggregate decreased the density.

The F-case specimens and S-case specimens presented the lowest and highest strain under the maximum stress respectively. The higher strain of R-case than that of F-case is mainly due to presence of interface between old cement mortar-aggregate and old cement mortar-SPB. Those interfaces in concrete may give rise to a progressive development of micro-cracks. The elastic modulus of sulfur concrete is generally lower than that of ordinary Portland cement concrete which has the same compressive strength with sulfur concrete. This means that sulfur concrete have bigger strain than ordinary Portland cement concrete under the same stress.

The average compressive strength of sulfur concrete was decreased 10 % after immersion of 60 days in acid and salt solutions (10 % HCl, 20 % H_2SO_4, 3 % NaCl). The F-case showed the most significant strength reductions as 17 %, while the R case presented the smallest strength reductions as 2 % in the three different solutions. The F-case and R-case showed 0.4 % of average mass reductions, while the S-case presented only 0.1 % of mass reductions in the three different solutions

The average of the measured coefficients of thermal expansion of sulfur concretes is bigger than that of Portland cement concrete. The coefficient of thermal expansion of sulfur concrete was reduced by mixing fly ash. It would be explained that SPB has higher coefficient of thermal expansion than that of fly ash. Using recycled coarse aggregate cause slight increase of coefficient of thermal expansion. It is expected that attached cement paste and mortar around recycled aggregate influenced increase of coefficient of thermal expansion of R-case.

The tested sulfur concretes presented high resistance to freezing and thawing. S- and F-case specimens made with natural aggregates had 84.6% of relative dynamic elastic modulus. However, R-type specimens made with recycled aggregates showed 77.6%.

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