International Journal of Engineering and Advanced Technology (IJEAT)
ISSN: 2249-8958 (Online), Volume-8 Issue-4, April 2019

Application of thermodynamic model to mix design of stabilized soils

Huu Nam Nguyen, Van Quan Tran, Anh Quan Ngo, Canh Tung Ngo

Abstract*: The purpose of finding suitable mixes for construction, sample experiments of each mix design of stabilized soils should be tested. However, in order to improve the accuracy of the experiment, the number of samples to be conducted is high, leading to high accuracy of the test results. The basic mechanisms are general so they have not really analyzed in detail the reclamation mechanism of basalt soil. The unspecified mineral mechanism will participate in the soil stabilization process. Based on the thermodynamic model, the paper focuses on explaining in detail the role of each mineral component of the soil for improving the mechanical properties of stabilized in soil. As well as the reaction mechanism of the binder mixture with the mineral components of the soil. The results of the model are compared relative to the experimental results to determine the correctness of the thermodynamic model as well as the results of the experiment, proving the feasibility of reinforcing basalt soil by mixture natural pozzolan, fly ash and cement. Mineral content of different mix designsis predicted by thermodynamic model thereby predicting the mechanical capacity of each mix design.

Index Terms*: Thermodynamic model, stabilized soils, mix design, natural pozzolan/lime/cement.

I. INTRODUCTION

Thermodynamics is essential for our understanding of chemical reactions. With the three most important variables, temperature, pressure and chemical composition, we can predict if the reaction will take place and the final state after the reaction ends. The general laws of thermodynamic regulation have long been known and were first applied to cement chemistry at the end of the 19th century by Le Chatelier to prove that the process of hydration of cement is obtained through the dissolution of clinker initially leads to an always saturated water phase for hydration reactions, leading to precipitation of solid phases. Finally, the remaining liquid solid phase equilibrium is reached in the pore system of the solid phase of cement. Since the 1940s, the period of research on applied thermodynamics has been strongly developed. Some things to give birth to the development of thermodynamic applications are the introduction of many fundamental laws, examples of thermodynamic characteristics of different phases: melting, flocculation, glass. Reflect on the relationship between balance and the reaction rate of substances.

The thermodynamic model is developed and applied by geochemists to calculate the complex equation system of polymorphic systems that occurs in nature. Thermodynamic models have been applied in many areas. For example, predicting the durability of underground construction of radioactive waste with cement / clay / radioactive interactions [1], [2], modelling interaction with clay or bentonite and pozzolan adhesives [3], [4]. It can be seen that the application of the interactive thermodynamic model in pozzolan environment is abundant. Therefore, the thermodynamic model can be applied to study the reaction mechanism between basalt soil - natural pozzolan - artificial pozzolan such as fly ash and cement. The purpose of finding suitable mixes for construction, sample experiments of each mix design of stabilized soils should be tested. However, in order to improve the accuracy of the experiment, the number of samples to be conducted is high, leading to high accuracy of the test results. The basic mechanisms are general so they have not really analyzed in detail the reclamation mechanism of basalt soil. The unspecified mineral mechanism will participate in the soil stabilization process. Based on the thermodynamic model, the paper focuses on explaining in detail the role of each mineral component of the soil for improving the mechanical properties of stabilized in soil. As well as the reaction mechanism of the binder mixture with the mineral components of the soil. The results of the model are compared relative to the experimental results to determine the correctness of the thermodynamic model as well as the results of the experiment, proving the feasibility of reinforcing basalt soil by mixture natural pozzolan, fly ash and cement. Mineral content of different mix designs is predicted by thermodynamic model thereby predicting the mechanical capacity of each mix design

II. MODELLING APROACH

A. Thermodynamic equilibrium

The interaction between the ionic species and the mineral species leads to precipitation/dissolution of minerals. The mineral saturation ratio Ω_m can be expressed as:

$$\Omega_m = K_{s,m}^{-1} \prod_{j=1}^{N_c} (\gamma_j C_j)^{\nu_{mj}} \qquad m = 1, \dots, N_p \qquad (1)$$

Where m is the indice of the mineral species, $K_{s,m}$ is the equilibrium constant.C_j is the molar concentration of primary species in the solution (mol.kg-1) i.e. the species that are supposed to move in the solution/ Other ions (often more complex) are taken into account when ionic complexations are calculated (see equation 3).

Manuscript published on 30 April 2019.
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ν_{mj} is the stoichiometric coefficient of the primary species, γ_j is the activity coefficient of the ion j , N_c, N_p are the number of corresponding primary species and mineral species. The state of equilibrium (or disequilibrium) of mineral species in the solution is controlled by the mineral saturation index IS_m, as follows:

$$IS_m = \log \Omega_m \qquad (2)$$

For a given mineral species, the solution is in equilibrium with the mineral species if $IS_m = 0$. The solution is under-saturated and the mineral species can still dissolve if $IS_m < 0$. Finally, the solution is super-saturated and the mineral species may be precipitated if $IS_m > 0$. Aqueous complexes are formed by interactions between primary species in the solution. These reactions are assumed to be at local equilibrium. By using the mass action law, the concentration of aqueous complexes can be expressed as a function of the concentration of primary species, as follows:

$$C_i = K_{c,i}^{-1} \gamma_i^{-1} \prod_{j=1}^{N_c} (\gamma_j C_j)^{\nu_{ij}} \qquad i = 1, \dots, N_x \qquad (3)$$

where C_i is the molal concentration of the aqueous complexes i ($mol.kg^{-1}$). γ_i, γ_j are the activity coefficients. $K_{c,i}$ is the equilibrium constant of aqueous complexation. N_x is the number of aqueous complexes considered in the solution. Thermodynamic database includes these parameters, the thermodynamic database THERMODDEM of Blanc et al [5] is applied in this paper.

B. Chemical reaction and required input data

Mineralogical composition of the initial materials: Soil, lime, volcanic ash was identified by powder X-ray diffraction (XRD). The Bogue calculation to determine the unhydrated clinker phases in the ordinary Portland cement (OPC) is more detailed in the Tran's work [6]. The initial mineralogical composition of the mixture is given in **Error! Reference source not found.**

Table 1.Mineralogical composition of the initial materials

Material	Phase	Formula	Composition (g/100g)
Soil	Quartz	SiO_2	4
	Kaolinite	$Al_2Si_2O_5(OH)_4$	23
	Gibbsite	$Al(OH)_3$	61
Lime	Lime	CaO	66
	Portlandite	$Ca(OH)_2$	16
	Calcite	$CaCO_3$	9
	Periclas	MgO	3
	Quartz	SiO_2	1
Natural pozzolan Daknong	Diopside	$CaMg(SiO_3)_2$	29
	Forsterite	Mg_2SiO_4	23
	Cristobalite	SiO_2	1
	Albite	$NaAlSi_3O_8$	27
	Quartz	SiO_2	4
Unhydrated Portland cement	Alite (C3S)	Ca_3SiO_5 or $(3CaO.SiO_2)$	16.75
	Belite (C2S)	Ca_2SiO_4 or $(2CaO.SiO_2)$	54.45
	Aluminate (C3A)	$Ca_3Al_2O_6$ or $(3CaO.Al_2O_3)$	14.08
	Ferrites (C4AF)	$Ca_4Al_2Fe_2O_{10}$ or $(4CaO.Al_2O_3.Fe_2O_3)$	8.52
	Gypsum	$CaSO_4.2H_2O$	1.40

The chemical reaction system of soil stabilization process is presented in the table 2, as well as the equilibrium constant at ambient environment 25°C, 1 atm

Table 2.Chemical reaction and equilibrium constant at 25°C, 1 atm.

Chemical reaction	
$Quartz + 2H_2O = H_4SiO_4$	-3.74
$Kaolinite + 6H^+ = 2Al^{3+} + 2H_4SiO_4 + H_2O$	6.47
$Gibbsite + 3H^+ = Al^{3+} + 3H_2O$	7.74
$Hematite + 6H^+ = 2Fe^{3+} + 3H_2O$	-0.06
$Goethite + 3H^+ = Fe^{3+} + 2H_2O$	0.36
$Lime + 2H^+ = Ca^{2+} + H_2O$	32.70
$Portlandite + 2H^+ = Ca^{2+} + 2H_2O$	22.81
$Calcite + H^+ = Ca^{2+} + HCO_3^-$	1.85
$Periclas + 2H^+ = Mg^{2+} + H_2O$	21.59
$Diopside + 4H^+ + 2H_2O = Ca^{2+} + Mg^{2+} + 2H_4SiO_4$	21.73
$Forsterite + 4H^+ = 2Mg^{2+} + H_4SiO_4$	28.60
$Cristobalite + 2H_2O = H_4SiO_4$	-3.16
$Albite + 4H^+ + 4H_2O = Al^{3+} + Na^+ + 3H_4SiO_4$	4.14
$C\text{-}S\text{-}H1.6 + 3.2H^+ = 1.6Ca^{2+} + H_4SiO_4 + 2.18H_2O$	28.00
$C\text{-}S\text{-}H1.2 + 2.4H^+ = 1.2Ca^{2+} + H_4SiO_4 + 1.26H_2O$	19.30
$C\text{-}S\text{-}H0.8 + 1.6H^+ = 0.8Ca^{2+} + H_4SiO_4 + 0.34H_2O$	11.05
$Monosulfoaluminate + 12H^+ = 2Al^{3+} + 4Ca^{2+} + SO_4^{2-} + 18H_2O$	73.09
$Ettringite + 12H^+ = 2Al^{3+} + 6Ca^{2+} + 3SO_4^{2-} + 38H_2O$	57.01
$Strätlingite + 10H^+ = 2Al^{3+} + 2Ca^{2+} + H_4SiO_4 + 10.5H_2O$	49.67
$Hydrotalcite + 14H^+ = 2Al^{3+} + 4Mg^{2+} + 17H_2O$	73.76
$Hydrogarnet + 12H^+ = 2Al^{3+} + 3Ca^{2+} + 12H_2O$	49.67
$Brucite + 2H^+ = Mg^{2+} + 2H_2O$	17.11
$Monocarboaluminate + 13H^+ = 2Al^{3+} + 4Ca^{2+} + HCO_3^- + 16.60H_2O$	80.54

C. Code Geochemical

The above reaction equations are stored in the response database. There are many reactive databases built to simulate thermodynamic balance simulations such as Phreeqc [7], Cemdata and Nagra-psi Kernel[8], etc. However, the database can be built. The most complete is the Thermodem database built by the French Republic Geological Research Laboratory (BRGM) [5], which includes a complete mineral system of hydration and minerals of majority Soil types exist in nature. Balance equations in the database require software or code to read and run the simulation. There are many free software widely available in practice such as GEMS-PSI of the Swiss geological agency, the code of Chess software developed by Paris University. However, it can be said that the most popular and easy-to-use software is Phreeqc built by the US geological agency Parkhurst and Appelo[7]. Thus all of thermodynamic equilibrium of the paper are carried out by Phreeqc code.

III. THERMODYNAMIC MODEL VALIDATED

In this section, the content of lime and cement is chosen as constant values, respectively 4% and 3% of the volume of stabilized soil in all simulations. The mix design of natural soil / pozzolan is simulated. The amount of stabilized soil is 100 g, the ratio of natural soil / natural pozzolan is changed according to the natural volume of pozzolan from 0 g, 5 g, 10 g, 15 g, 20 g, 25 g, 30 g and 40 g. Numerical results are compared with experimental results which is extracted from Vu et al. [10].The comparison is presented in Figure 1

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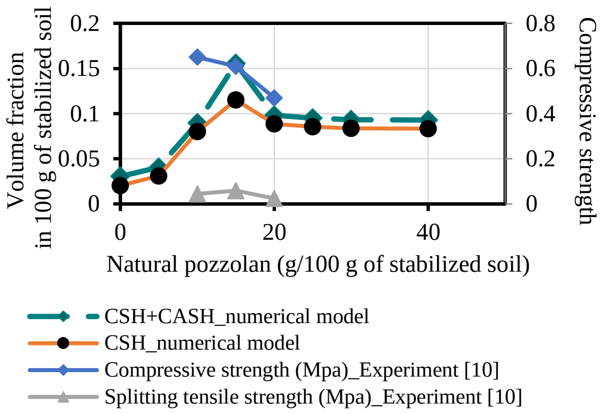

Figure1.Amount of C-S-H, C-S-H+C-A-S-H vs Compressive strength and splitting tensile [10]

Through the model results, the calcium silicate content (C-S-H) and the total C-S-H + C-A-S-H content according to the amount of natural pozzolan used for soil stabilization can be described through two regions. Region one, C-S-H or C-S-H + C-A-S-H content increases in proportion to the increase in the amount of pozzolan used. C-S-H calcium silicate content, C-S-H + C-A-S-H is most formed in stabilized soil when the amount of natural pozzolan used is 15% (or 15 g per 100 g of stabilized soil) with 83% natural soil, 4 % lime and 3% cement. After achieving the highest content, C-S-H, C-S-H + C-A-S-H content formed gradually, although natural pozzolan content continued to increase in the second region. At the same time, the results of the experiment showed that the behavior of the strength of the splitting soil of the stabilized soil depends on the amount of natural pozzolan used, this behavior corresponds to the change of C-S-H calcium silicate content, or C-S-H + C-A-S-H. The development of compressive strength has not really corresponded with the development of C-S-H content, or C-S-H + C-A-S-H according to the amount of natural pozzolan used, but it can be seen that the amount of these glue together with the intensity compression pull is reduced when the natural pozzolan content is greater than 15%. It is known that C-S-H calcium silicate mineral is the largest mineral contributing to the strength of cement materials (compressive strength, strength of splitting ...) [11]. Therefore, mechanical properties development of stabilized soil depends largely on the C-S-H content. The results of the model show that the thermodynamic model seems to be relatively approximate to the development of the strength of stabilized soil according to the amount of pozzolan used, hence the thermodynamic model can be used as an initial dè design tool to find the optimal mix when using natural pozzolan for stabilized soil.

Further analysis of the composition of the remaining minerals after achieving equilibrium of the mixture of stabilized soil materials with the grade of "natural soil / pozzolan / cement / lime = 78/15/3/4" by The result of the standardized chemical thermodynamic model of minerals by percentage is shown in the following table Table 3:

Table 3.Standardized content of minerals in the stabilized soil after equilibrium reached

Material	Phase	Formula	Standardized mineral amount (%)
Soil	Quartz	SiO_2	100
	Kaolinite	$Al_2Si_2O_5(OH)_4$	100
	Gibbsite	$Al(OH)_3$	56
Lime	Lime	CaO	0
	Portlandite	$Ca(OH)_2$	100
	Calcite	$CaCO_3$	100
	Periclas	MgO	0
	Quartz	SiO_2	100
Natural pozzolan	Diopside	$CaMg(SiO_3)_2$	100
	Forsterite	Mg_2SiO_4	0
	Cristobalite	SiO_2	0
	Albite	$NaAlSi_3O_8$	100
	Quartz	SiO_2	100
Unhydrated Portland cement	Alite	Ca_3SiO_5or ($3CaO.SiO_2$)	0
	Belite	Ca_2SiO_4or ($2CaO. SiO_2$)	0
	Aluminate	$Ca_3Al_2O_6$ or ($3CaO.Al_2O_3$)	0
	Ferrites	$Ca_4Al_2Fe_2O_{10}$ or ($4CaO.Al_2O_3.Fe_2O_3$)	0

Through the results in table 3, it can be seen that natural lime and pozzolan and cement have great activity when free lime and cement minerals completely react to produce the main components of C-S-H and C-A-S-H. With natural pozzolan, it is easy to see that the natural activity of pozzolan depends on the content of Forsterite and Cristobalite, when these two minerals fully react to produce soluble molecules $H_4SiO_{4(aqueous)}$ in solution. Soluble molecule $H_4SiO_{4(aqueous)}$ can be written as the following equation:

$$H_4SiO_{4(aqueous)} = SiO_{2(aqueous)} + H_2O \qquad (4)$$

Natural soil participates in the stabilization process by dissolving the gibbsite mineral to produce Al + 3 ions involved in the creation of C-A-S-H (Strätlingite). Free Ca + 2 ions are provided by CaO free lime and cement minerals.

In addition to checking the correctness of the model, model results and experimental results in the case of comparing the role of lime in soil stabilization, the distribution uses 10% natural pozzolan, 3% Cement and 4% lime are compared with similar soil mixes, natural pozzolan, cement and lime free.

Table 4.Experimental compressive strength of sample after 14 days in saturation conditions and mineral content of C-S-H and C-A-S-H simulated in two mix design of stabilized soil using lime and not using lime

Lime content (g/100g)	Experiment	Numerical (Volume fraction)		
	Average compressive strength of 3 samples (MPa)	C-S-H	C-A-S-H	C-S-H+ C-A-S-H
0	0.33667	0.04371	0.00954	0.05325
4	0.65000	0.08004	0.00954	0.08958

The results presented in table 4 show that using 10% natural pozzolan and 3% cement, the lime content has an effect on the compressive strength of the test.

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With 4% lime content used, the compressive strength of stabilized soil almost doubles. The results of the experimental compressive strength are controlled with the results of the numerical model. The numerical model shows constant C-A-S-H mineral content, however, the C-S-H and mineral content provides the main strength for cement materials, almost doubling in proportion to the double compressive strength. C-S-H content increased due to free CaO lime in dissolved lime created Ca^{+2}, causing imbalance of the reaction system and complete dissolution of Forsterite to create soluble molecules $H_4SiO_{4(aqueous)}$ $(SiO_{2(aqueous)})$ to combine Ca^{+2} ions to produce C-S-H (C-S-H1.6, C-S-H1.2 and C-S-H0.8), this mechanism does not appear in the field. The combination does not use lime, which leads to very small dissolution of Forsterite mineral. Since the activity of natural pozzolan is not maximized. From this, it can be seen that the important role of lime in activating the activity of natural pozzolan in stabilized soil process.

Experimental results and numerical models are again controlled, showing that the correctness of the thermodynamic model is designed to study the reaction mechanism of substances in a mixture of soil stabilization materials. In order to understand and compare the role of substances in mixes if stabilized soil, the paper simulates and compares with the experimental results of two mix design: soil-cement-natural pozzolan and soil-cement. in the next section.

IV. Thermodynamic Equilibrium of Soil-Cement-Natural Pozzolan

In this section, the thermodynamic model will study the activation capacity of cement for Daknong natural pozzolan in the process of reinforcing unused lime. The result of the model, which is the mineral content that gives mechanical capability to C-S-H and C-A-S-H materials, will be controlled with the results of the test: compressive strength and splitting tensile strengthof specimen cured in saturated conditions after 14 days of age. The two mixes design used for the study are "soil / natural pozzolan / cement = 90/0/10 and 80/10/10". Results of experimental compressive strength of sample after 14 days in saturation conditions and numerical C-S-H and C-A-S-H mineral content of two mixes design of stabilized soil using natural pozzolan and not using natural pozzolan presented in table 5

Table 5. Experimental compressive strength of the sample after 14 days in saturation conditions and the simulated mineral content of C-S-H and C-A-S-H of two mixes design of stabilized soil using natural pozzolan and not using natural pozzolan

Mix design	P/C=0/10	P/C=10/10
Compressive strength (MPa)	1.1	1.07
Splitting tensile strength (MPa)	0.085	0.086
C-S-H (volume fraction)	0.0725	0.0735
C-A-S-H (volume fraction)	0.0239	0.0239
C-S-H+C-A-S-H (volume fraction)	0.0964	0.0973

Through the results in the table above it can be seen that the content of C-S-H and C-A-S-H minerals is relatively equivalent in two cases of natural pozzolan and no natural pozzolan with cement as an activator. This may explain that activators such as CaO free in cement are only sufficiently used primarily for hydration reactions of cement, which is evident when CaO oxides exist under properties. mineral products C_3S, C_2S, C_3A, C_4AF. Therefore, the mineral components of Forsterite and Cristobalite natural pozzolan help pozzolanic reactions to take place, but in the case of using cement as activator, only Cristobalite with very small content is dissolved, so the difference The difference in CSH mineral content in the case of using natural pozzolan and not using natural pozzolan is relatively small. Therefore, when using cement without lime as a trigger for Daknong's natural pozzolan, it is not effective to reinforce soil. At the same time, the results of the C-S-H and C-A-S-H mineral content of the model are relatively consistent with the mechanical strength of the stabilized soil sample. Through the above two sections III and IV, the relative accuracy of the thermodynamic model has been confirmed in order to further investigate the mechanism of the soil stabilization process using Dankong natural pozzolan, cement and lime.

V. Application of Thermodynamic Model to Optimize Mix Design

In order to find the optimal mix of natural soil / lime / cement / pozzolan, the content of each component will be changed. Lime has three contents of 0%, 4% and 10% by weight of the mixture. Cement also has three levels of 0%, 3% and 10%. Natural pozzolan has eight contents of 0%, 5%, 10%, 15%, 20%, 30% and 40%. The percentage content of used soil mass is changed so that the total volume of mixed materials reaches 100gram. These mixed materials are in turn equilibrated by thermodynamic models to determine C-S-H calcium silicate mineral content and silicate calcium aluminate C-A-S-H. Thus, the number of mixes design is simulated by the thermodynamic model of 3 x 3 x 7 = 63 mixes. The results of the model are shown in Figure 2, Figure 3, and Figure 4 below. As a result, the C-S-H and C-A-S-H contents are in 100 g of the soil stabilization mixture. This content depends on the percentage content of natural pozzolan, lime and cement. Optimal mix with lime used 0%, 4% and 10% respectively "natural cement / pozzolan = 10/15; 10/20 and 10/20". C-S-H + C-A-S-H content is greatest when the ratio of natural lime / cement / pozzolanstabilization is 10/10/20. When lime and cement are not used, the stabilized soil mixture does not produce C-S-H + C-A-S-H despite the presence of natural pozzolan

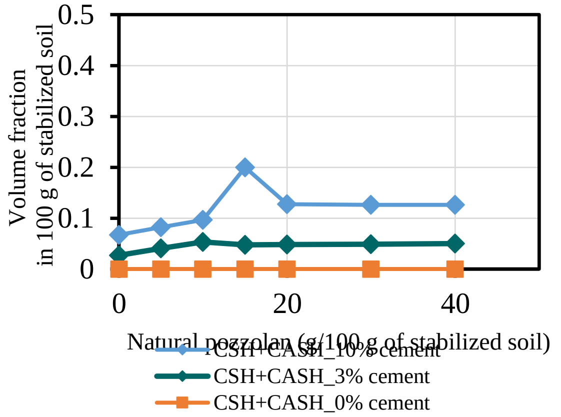

Figure 2.Numerical results of mineral content C-S-H + C-A-S-H in 100 g of stabilizedsoil in percentage of natural pozzolan, cement and 0% lime.

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When cement is not used in the stabilization mixture, the C S-H+C-A-S-H content or the strenth increases proportionately according to the natural pozzolan content. At the same time, the results also showed that if using natural lime and pozzolan as the only material to reinforce the soil, CS-H + CASH content is relatively small at 3% lime or 10% lime, so if only used lime as a trigger for pozzolan reactions, the strength of the stabilized soil does not thrive despite the heavy use of lime, which is consistent with the soil stabilization design of the Indiana state geotechnical centre of United State [12].

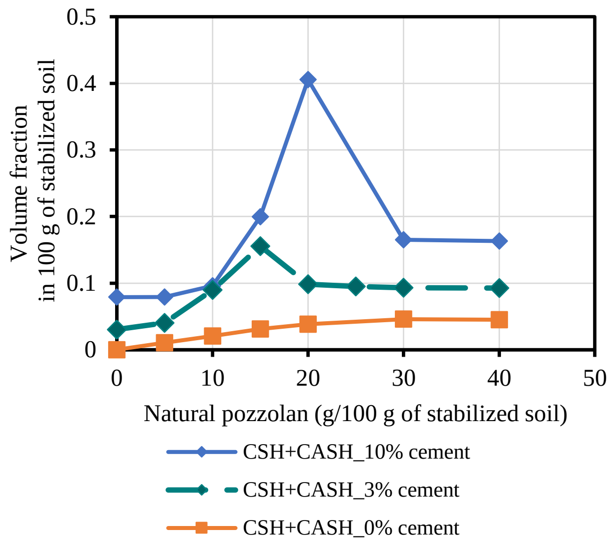

Figure 3.Numerical results of mineral content C-S-H + C-A-S-H in 100 g of stabilizedsoil in percentage of natural pozzolan, cement and 4% lime.

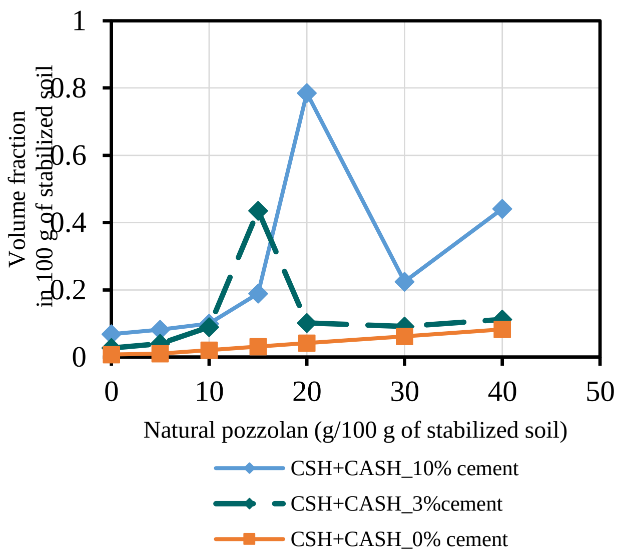

Figure 4.Numerical results of mineral content C-S-H + C-A-S-H in 100 g of stabilizedsoil in percentage of natural pozzolan, cement and 10% lime.

VI. CONCLUSIONS

The paper has successfully built a thermodynamic model to explain the mechanism of soil stabilization with adhesives: natural lime, cement and natural pozzolan. The theory of soil stabilization mechanism has been reviewed in a generalized way since then the thermodynamic model has been proposed to further study the stabilization mechanism. The basic theory of equilibrium thermodynamic models, as well as the applicability of models in material science has been presented. By controlling the results of the strength experimentof mixes design “soil/natural pozzolan/lime/cement”, “soil /natural pozzolan/cement” and “soil/cement”, the experimental results verified the correctness of the model.

The model has a relatively high accuracy, which is applicable as a support tool to design soil stabilization using natural pozzolan, lime and cement. Thereby reducing the design cost by reducing the number of samples, and checking each other of the accuracy of test results and models, thereby improving the digital model. Due to the limitation of the paper, many research issues of the soil stabilization mechanism have not been clarified and integrated thermodynamic models such as the influence of temperature, the effect of pH, the role of cement. in the reaction process with different lime content. At the same time the time of the reaction from that predicts the development of intensity over time. These issues suggest the paper needs simulation studies combined with experiments to improve accuracy and expand the simulation ability of thermodynamic models in other studies in the future.

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Blue Eyes Intelligence Engineering
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