ON WEATHERING AND CHANGE OF PROPERTIES OF TERTIARY MUDSTONE RELATED TO LANDSLIDE

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SYNOPSIS

During the course of study on the mechanism of the deterioration of bedrock (mudstone) in landslide zones, some facts were found to suggest that the softening of mudstone is mainly due to the amount of Gibbs' free energy evolved when hydrogen-bond is formed between originally adsorbed water molecules and newly adsorbed ones around clay particles.

It was also found that the relationship that $q=\mathrm{A}\ \exp\mathrm{B}\ (\gamma_t\text{-}\gamma_{t_0})$ which was already confirmed concerning clay soils can be applied to mudstone if the former is originated from the latter, though there is a sharp difference in the water-resistibility between them.

1. *Introduction*

Landslides occurring on Tertiary sedimentary rocks (mudstone or shale) are of great importance in Japan where deposits in that period are exposed extensively.

The bedrock or mudstone in such landslide areas being weathered and gradually weakened into landslide clay in course of time, it is necessary, especially in the study

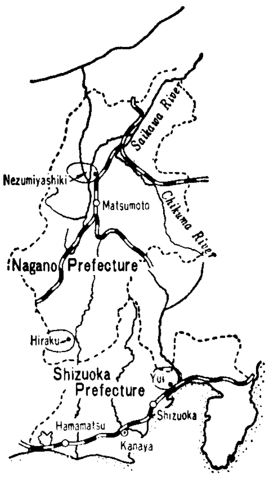

Fig. 1. Locality of landslides investigated

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Table 1. Locality, Geology and Gradient of Sites

	Locality of landslide	Geology	mean gradient of slope
1	Imajuku, Yuimachi, Shizuoka Pref.	Pliocene	10°~14°
2	Hiraku, Anancho, Nagano Pref.	Miocene	15°~20°
3	Nezumiyashiki, Honjomura, Nagano Pref.	Miocene	17°~22°

Note: For convenience, sake, locality 1 is expressed as Yui, 2 as Hiraku and 3 as Nezumiyashiki, respectively.

of long-term stability problems of slopes, to clarify the mechanism of the process of weathering and the change of physical and mechanical properties of bedrocks accompanying the softening process. An investigation was then made on Tertiary mudstones sampled from three landslide areas, Yui, Hiraku, and Nezumiyashiki by name. (see Table 1 and Fig. 1).

For details about the landslide at Yui, refer to the reference 1).

2. *Definition of clay soil and mudstone*

In view of water-resistibility, clay soil and mudstone are defined respectively as follows:

1. Clay soil: material which is composed mainly of clay-fraction and is easily weakened, reducing its strength almost to zero, when the material with natural water content is immersed in water.

2. Mudstone: material which in the same case keeps its original state and whose strength hardly decreases.

When immersed in water, a mudstone also swells slowly, decreasing in its bulk density and strength. The rate of reduction of the bulk density is, however, far slower than that of clay soil.

The unconfined compression strength of a rather fresh natural mudstone obtained at the bottom of a valley stream at Yui, for example, is 20–40 kg/cm^2 and that of identical mudstone left in water for one year is 10 kg/cm^2. In other words, the mudstone still remains considerably hard. Though both clay soil and mudstone are almost fully saturated in-situ, they swell when immersed in water. This means that the water invading the pores of clay or mudstone is neither capillary nor gravity water but is chemically weakly bound water[1)] which can be drained out neither by underdrains nor by horizontal boreholes.

3. *On the mechanism of the softening of mudstone*

3.1. Mechanism of softening of dried mudstone by slaking

As mentioned, above mudstone with natural water content does not disintegrate, even after being immersed in water for a long time. If, however, once dried and then wetted, it rapidly disintegrates into small pieces, or slakes. In order to find a clue to the solution of the mechanism of this phenomenon, some experiments were performed as described below.

3.1.1. Comparison of slaking in atmospheric pressure and in vacuum.

If slaking is mainly due to the tension produced by trapped air, as Terzaghi explained, there must be a clear difference between the slaking in vacuum and in the air.

In order to ascertain this, a block of dried mudstone was divided into two parts, one was immersed in water in atmospheric pressure and the other in vacuum (15 mmHg) (the latter was left in vacuum for two hours before immersed in water).

In both cases, the mudstone rapidly disintegrated into small pieces and no substantial difference was observed between the mudstone slaked in the air and that in vacuum, though the percentage of the smaller grains of the former is a little larger than that of the latter.

On the basis of this observation, the author assumed that slaking of dried mudstone is not mainly due to the tension produced by trapped air, but to some physico-chemical action, and the following investigation was undertaken.

3.1.2. Reaction of air-dried mudstone against water and organic liquids

Air-dried mudstone was immersed in water and organic liquids as was previously tested by N. Y. Denisov and B. F. Reltov.[3)]

The results are shown in Photographs 1～2 and Table 2.

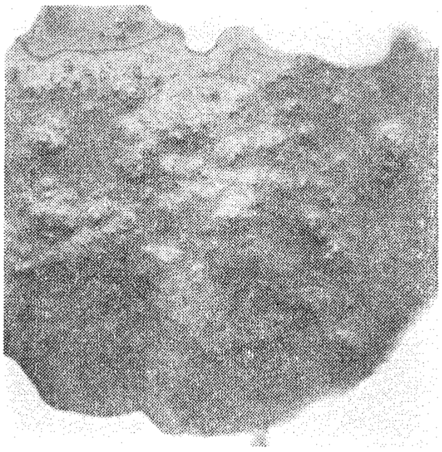

Photo 1 Mudstone slaked in water (intensity of reaction in Table 2: 5, sample: Yui)

Photo 2 Mudstone slaked in methanol (intensity of reaction in Table 2: 3, sample: Yui)

3.1.3. Considerations

Denisov and Reltov pointed out that the destructive effect of liquids depends upon the value of the dielectric constant. The results in Table 2 show, however, that the destructive effect does not necessarily depend upon the dielectric constant (see, e.g. the case of nitrobenzene).

Since the value of the dielectric constant is no more than a physico-chemical index of any substance microscopically the fact that the degree of slaking does not necessarily depend upon the value means that it is necessary to consider this problem microscopically i.e. from the standpoint of chemical bond.

Table 2. Reaction of Air-dried Mudstone against Water and Organic Liquids

Name of liquids	Chemical formula	Dielectric constant c.g.s.e.s.u.	Solubility in water	Intensity of reaction			Note	
				Yui	Hiraku	Nezumi-yashiki	Surface tension (contact with air) dyne/cm	Coefficient of viscosity poise
water	H_2O	81	∞	5	5	4+	73	0.011
glycerine	HO_2CH_2-CHOH $\cdot CH_2OH$	43	∞	4	2	2	63	15~40
ethylene-glycol	$CH_2OH \cdot CH_2OH$	39	∞	4	1	2	48	0.2
nitro-benzene	$C_6H_5 \cdot NO_2$	36	0.2	0	0	0	44	0.02
methanol	$CH_3 \cdot OH$	33	∞	3	1	2	23	0.006
ethanol	$C_2H_5 \cdot OH$	26	∞	2	0	1−	22	0.01
propanol	$CH_3(CH_2)_2 \cdot OH$	22	∞	1	0	1−−	24	0.02
acetone	$CH_3 \cdot CO \cdot CH_3$	21	∞	4+	1+	2	24	0.004
butanol	$CH_3(CH_2)_3 \cdot OH$	19	7.4	1−	0	0	25	0.03
methyl ethyl ketone	$CH_3 \cdot CO \cdot C_2H_5$	19	33.5	3	1−	1+	25	0.004
diethyl ketone	$C_2H_5 \cdot CO \cdot C_2H_5$	17	4.2	0	0	0	25	0.005
ethyl ether	$C_2H_5 \cdot O \cdot C_2H_5$	4	7.5	0	0	0	22	0.002
xylene	$C_6H_4(CH_3)_2$	m.2 p.2 0.2	0.008	0	0	0	29 28 30	0.009 0.007 0.007
carbon tetrachloride	CCl_4	2	0.08	0	0	0	27	0.01
benzene	C_6H_6	2	0.06	0	0	0	29	0.007
liquid paraffin	C_nH_{2n+2}	2	0.003	0	0	0	33*	3~4

Note :

① The liquids are arranged in the order of dielectric constants.

② Among the surface tensions, that with an asterisk(*) was measured by the author with De Nouy tensiometer, while other constants are from literatures.

③ The "solubility in water" is expressed by the number in grams of dissolved qantity of liquid in 100 grams of water. The mark "∞" means that the liquid dissolves in water in any rate.

④ No reaction is expressed as zero, while the intensity of reaction is expressed in terms of five point scale according to the observation of the degree of destruction when the reaction stops. Those mudstones immersed in high viscosity liquids were observed after seperated into pieces by slight shaking, lest the degree of reaction should be underestimated, because those pieces do not separate in spite of cracks.

From such a point of view, the liquids reacted with mudstone were water, alchohol, and ketone and they are able to form close hydrogen-bonds with water molecules: from the data already published, it is assumed that water and alcohol molecules form hydrogen-bond with water molecules as proton-donors and concurrently as proton-accepter, while ketone solely as proton-accepter as shown in Fig. 2.

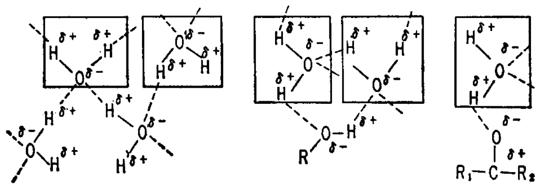

Fig. 2. (Left) Hydrogen-bond between water molecules (middle) Hydrogen-bond between alcohol (R.OH) and water molecules (Right Hydrogen-bond between ketone (R_1-O-R_2) and water molecules

Note:

① **Hydrogen-bond is shown by . . .**
② δ^+ **and** δ^- **show the polarized electric charge**
③ **Water molecules within ▭ express water adsorbed around clay particles before reaction**
④ **R shows alkyl-group**

Among these, for example, in saturated monohydroxy-alchohol group, the smaller the number of carbon is, the greater is the dielectric constant,[4] and consequently, the greater is the ability to form hydrogen-bond with water molecules which are originally adsorbed around clay particles of dried mudstone: therefore, the greater is the solubility in water.

In case of nitro-benzene, however, the ability to form hydrogen-bond with water molecules is very poor, and consequently, the solubility in water is small, though its dielectric constant is relatively large.

In view of the above facts, the most reasonable conclusion to be drawn is that the greater the ability of liquid to form hydrogen-bond with water molecules is, the more is the ability to destruct dried mudstone; the slaking is a kind of chemical dissolution due to hydrogen-bond.

When the network of molecules is formed by such hydrogen-bond, the chemical potential of water, alcohol and ketone becomes lower than before, and the excessive free energy is released.

Therefore, it may also be reasonable to assume that a part of this energy is used for the work of increasing surfaces of mudstone (i.e. the work of destruction by slaking).

This may be explained otherwise as follows: when polar molecules are held around clay particles through hydrogen-bond, the basal spacing of montmorillonite increases, causing strain in mudstone and destructing it. (It was already confirmed in the authors' laboratory that the mudstones dealt with in this paper contains, more or less, montmorillonite.)

3.2. On mechanism of the softening process of natural and artificial mudstones

3.2.1. Swelling of natural and artificial mudstones in water

As has already been described in Chapter 2, the mudstone with natural water content cannot be disintegrated even after being immersed in water for a long time, though it very slowly swells and its bulk density decreases a little.

If once dried, or if crushed not so as to make any change of water content and again compressed in a mold to approximately the same bulk density as that of natural mudstone (this is referred to as " artificial mudstone " in this paper for convenience, sake,

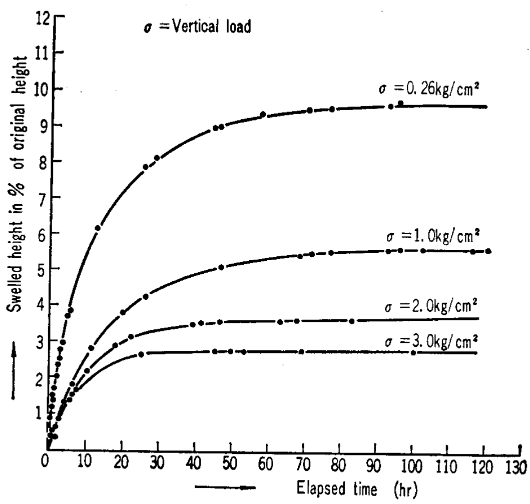

Fig. 3. Time-swelling curves of artificial mudstones in water under various vertical pressures (Laterally-confined, sample: Yui, γ_t before swelling: 2.26±0.01)

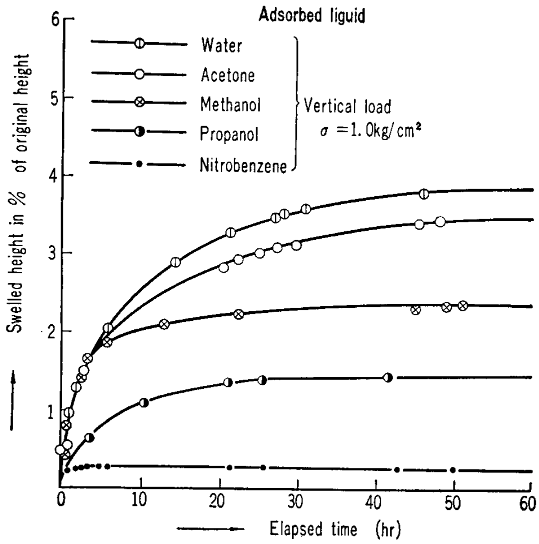

Fig. 4. Time-swelling curves of artificial mudstones under a vertical pressure of 1.0 kg/cm² in water and organic liquids. (Sample: Yui, laterally-confined. γ_t before swellihg: 2.22±0.02)

though the name contradicts the aforesaid definition of mudstone), however, it rapidly disintegrates and swells in water.

The time-swelling curves of artificial mudstone in water are shown in Fig. 3.

3.2.2. Swelling of artificial mudstone in various liquids.

Artificial mudstone also swells in some liquids other than water. The time-swelling curves of the artificial mudstone from Yui, under a vertical pressure of 1 kg/cm^2, in several liquids are shown in Fig. 4.

3.2.3. Considerations

As Fig. 4 shows, the order of the swelled volume is approximately in the same order as the intensity of slaking shown in Table 2, which may mean that the softening of natural mudstone is a similar phenomenon to the slaking of dried mudstone, or is a kind of chemical dissolution. In this case, it may also be reasonable to assume that a part of free energy evolved when the molecules of water, alchohol or ketone form a network due to hydrogen-bond play a role in making the mudstone swell.

From the experimental results described above, it may be clear that the chemical potential of the water adsorbed around clay particles in natural mudstone is considerably lower than that of free water. Nevertheless, natural mudstone hardly swells when in contact with free water.

This is probably why, judging from conclusions of some studies already reported,[3),5)] a substance like silicic acid gel forms a film making the water molecules around clay particles inactive.

Therefore, it may be reasonable to consider that, when there occurs a crack in this film and the active surface is exposed by " drying " and " crushing ", hydrogen-bonds are easily formed between originally absorbed water molecules and free water molecules, resulting in swelling and slaking.

3.3. Thermodynamical study on slaking of mudstone equilibrated in various relative humidities.

As previously mentioned, the mudstone is easily slaked and disintegrated by cyclic drying and wetting.

According to the data reported by Yamaguchi and others[6)], the velocity of seismic waves in geologically fractured zones in Kamenose landslide area in Osaka Prefecture was 1.5 km/sec. in dry season and 1.2 km/sec. in wet season, this fact may mean that the fluctuation of vapor pressure or water content in the ground is not negligible, especially in geologically fractured zones. In this connection, it would be of practical significance to investigate further to what extent the mudstone should be dried in order to make it slake.

3.3.1. Method of experiment and results

The mudstone cut by a diamond-cutter was dried in various relative humidities (from 98% to 0%) by desiccator-method and then some samples were immersed in water and others were tested to know their unconfined compressive strength q_u.

In addition, exampes of water content before and after slaking are shown in Table 3, as to mudstone equilibrated in 80% and 98% of relative humidity. The water

Table 3. Water Contents of Mudstones Equilibrated in 80% and 98% of Relative Humidities Before and After Slaking in Water.

Sample	Yui	
Relative humidity at equilibrium (%)	Water content of mudstone before slaking (%)	Water content of mudstone after slaking (%)
80	5.3±0.8	17.0±1.4
98	8.0±0.5	12.7±1.0

Note: Confidence interval of population mean of water content was calculated taking confidence-coefficient at 0.9.

contents shown in this Table were measured after the excessive water around the disintegrated pieces was slightly wiped off by gauze.

3.3.2. Considerations

As a result, it was found that drying in 98% of relative humidity is enough to make the mudstone start to slake in case of Yui and Hiraku, while in case of Nezumiyashiki drying in 94% of relative humidity is necessary for the mudstone to slake and observation also shows that in each case, the drier is the mudstone as compared with its natural state, the greater is the intensity of its disintegration in water.

Therefore the mudstone may easily be deteriorated especially within the zone of fluctuating water vapor pressure or fluctuating groundwater level. Since the change of vapor pressure is presumed to be small in the in-situ condition, the following thermodynamical consideration was made on the energies when relative humidity changes between 98% and 100% taking the case of mudstone of Yui for example.

① The chemical potential of water vapor in 98% of relative humidity

Denoting the chemical potential of 1 mol of water vapor in 100% of relative humidity (i.e. the chemical potential of pure free water) as G_1, $f^0_{H_2O}$ as fugacity, G_2 as the chemical potential of 1 mol of vapor in 98% of relative humidity and f_{H_2O} as its fugacity, the difference between G_1 and G_2 is given by the formula:

$$\Delta G = G_2 - G_1 = RT \ln \frac{f_{H_2O}}{f^0_{H_2O}} = 8.3 \times 10^7 \times 288 \times 2.30 \log_{10} 0.98 = -4.9 \times 10^8 \text{ erg/mol}$$

Here, R is the gas constant 8.3×10^7 erg/mol.

T is absolute temperature at 15°C (room temperature), 273+15=288°K, and the fugacity of vapor is taken to be nearly equal to the vapor pressure since a gas of low pressure can be regarded as an ideal gas.

② The "specific deformation energy" necessary for the destruction of mudstone by compression

The unconfined compressive strength of a mudstone equilibrated in the vapor (E) pressure of 98% of relative humidity is 50–80 kg/cm², and its modulus of elasticity is $2 \times 10^9 \sim 3 \times 10^9$ dyne/cm².

Assuming that the mudstone is approximately an isotropic elastic body with the

Poisson's ratio (μ=0.3), the "specific deformation energy"[7] necessary for the destruction of mudstone by compression is calculated by the formula:

$$V_f = \frac{1+\mu}{2E}\left(q_{u\,max} - \frac{q_{u\,max}}{3}\right)^2 + \left(\frac{q_{u\,max}}{3}\right)^2 + \left(\frac{q_{u\,max}}{3}\right)^2 = 6\times10^5 \sim 9\times10^5\ \mathrm{erg/cm^3}$$

③ An approximate value of the Gibbs' free energy released by water adsorbed in process of slaking

As shown in Table 3, the water content of a mudstone equilibrated in the vapor of 98% of relative humidity is about 8% on an average, and the water content of blocks produced by slaking is about 13% on an average.

Since the weight per unit volume of dried mudstone is 2.1 g/cm³ and the molecular weight of water being equal to 18, the moisture equivalent to 1% of water content is $2.1 \times 1/100 \times 1/18 = 1.2 \times 10^{-3}$ mol/cm³.

Considering that the chemical potential of adsorbed water (chemically weakly bound water) changes approximately linearly with water content as shown in Fig. 5, the

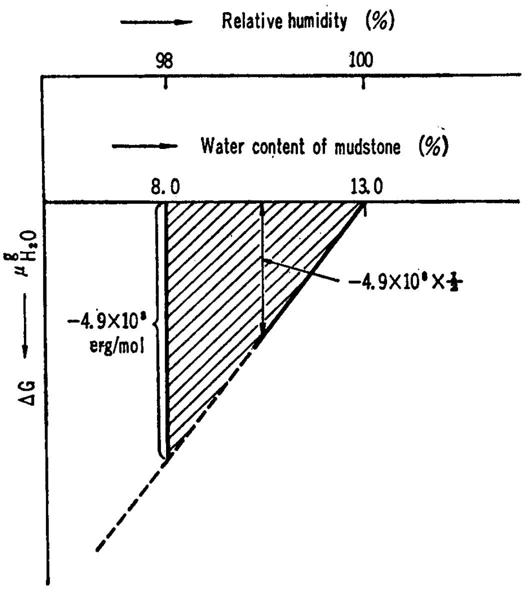

Fig. 5. The relation of water content of a mudstone to relative humidity and free energy

Note:

① $\mu^g_{H_2O}$ = **chemical potential of pure water in gas-phase of unit pressure**
= chemical potential of pure free water

② ΔG = **difference between chemical potential of 1 mol of pure free water and that of 1 mol of vapour in various relative humidities**

③ **shaded area corresponds to ΔE, i.e. Gibbs' free energy evolved by dried Yui-mudstone when its water content changes from 8% to 13% in water**

Gibbs' free energy ΔE released when the water content increases in the process of slaking is calculated as follows:

$$\Delta E = 4.9\times10^{8}\times1.2\times10^{-8}\times(13-8)\times\frac{1}{2} = 1.5\times10^{6}\ \mathrm{erg/cm^{3}}\ .$$

Thus, V_f is found to be 40–60% of ΔE, which means that twice or three times as much free energy as is necessary for the mechanical destruction of Yui mudstone are evolved with the fluctuation of vapor pressure only between 98% and 100% of relative humidity.

In an extreme case when mudstone does not slake at all, landslide may not occur even if its bedrock is the mudstone deposited in Tertiary period.

In fact, according to an investigation shown in Table 4, landslides do not occur at all or if they do very seldom on the formation (Bessho, Uchimura and Moriya formations) whose mudstones do not slake or, even if slake they do very little.

Table 4. Relation between Geology and Recorded Numbers of Landslides so far Occurred along Chikuma and Saikawa River Systems in Nagano Prefecture

Formation	Numbers of landslides
Sarumaru formation	11
Shigarami formation	45
Ogawa formation	98
Aoki formation	29
Bessho formation	2
Uchimura formation	0
Moriya formation	0
Sum total	185

Note : From up to down, the formations become older. Mudstone of Nezumiyashiki belongs to Aoki formation and that of Hiraku to Tomikusa formation which is of the same age as Aoki formation.

So far, considerations were made mainly of a comparatively shallow or near-surface part.

Even in deeper part where no fluctuation of vapor pressure or groundwater level occurs, softening of mudstone may inevitably proceed till the chemical potential of the adsorbed water around clay particles becomes equal to that of ground-water as the active surfaces of clay particles of mudstone are exposed and water molecules are adsorbed.

The active surface is presumably much exposed especially in the zone which has suffered intense geological fracturing.

Therefore, in view of the swelling of artificial mudstone, the rate of softening of mudstone in such a zone may not necessarily be slower.

Apart from such an intensely fractured zone, however, the rate of softening in deeper part may, in general, be slower since the silicic acid gel film (which is thought to inactivate the clay particles of mudstone) is difficult to dissolve in water and the difference between chemical potential of the adsorbed water and that of free water becomes smaller as the overburden pressure increases: thus, it seems reasonable to assume that the primary sliding surfaces or sliding zones are most likely to grow where groundwater or vapor pressure fluctuates and that they grow slower, though inevitably, in deeper part, and it might be concluded that the real cause of landslide often occurring in Tertiary mudstones in Japan is the softening of bedrocks due to the mechanism so far described and heavy rain or thawing of snow plays a role only of a trigger action.[8]

4. *On physical and mechanical properties of mudstone accompanying the softening process*

Weakening of mudstone proceeds with a mechanism previously described and phy-

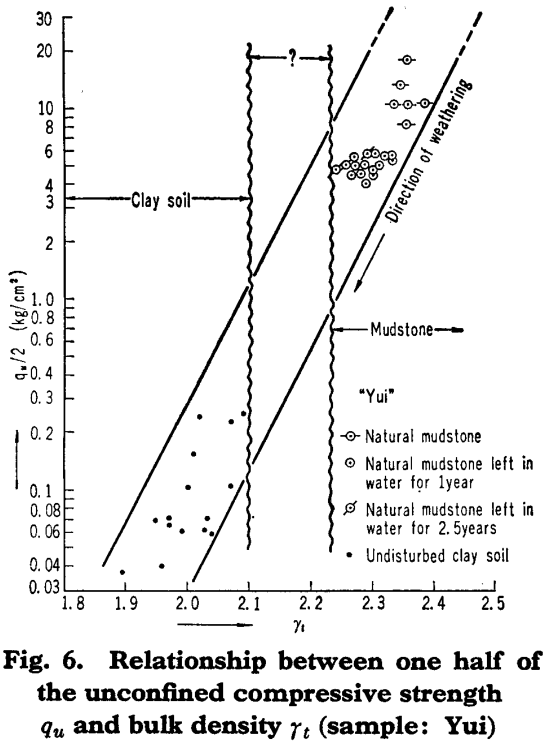

Fig. 6. Relationship between one half of the unconfined compressive strength q_u and bulk density γ_t (sample: Yui)

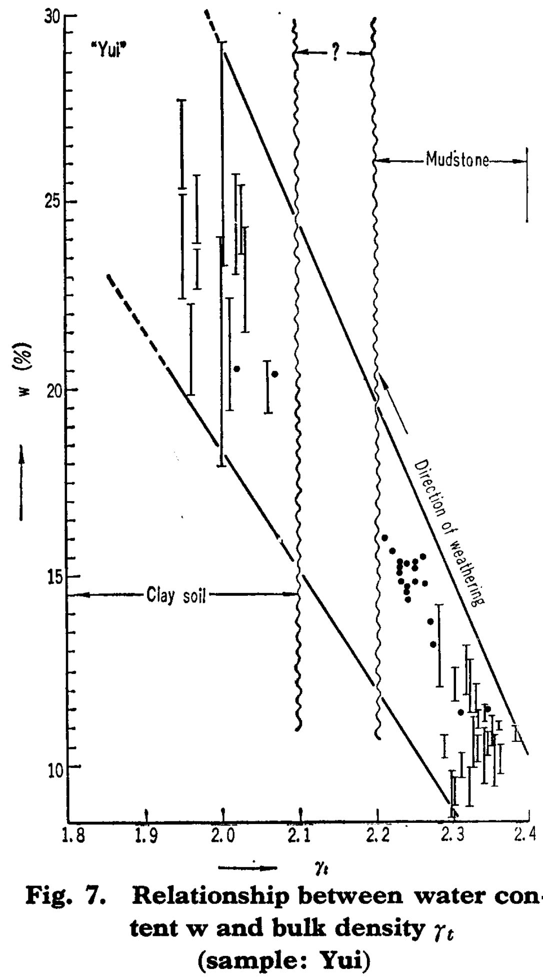

Fig. 7. Relationship between water content w and bulk density γ_t (sample: Yui)

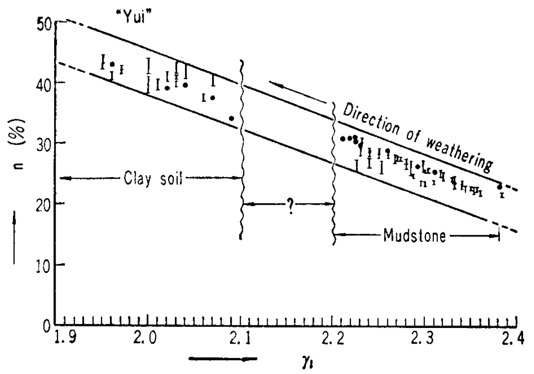

Fig. 8. Relationship between porosity n and bulk density γ_t (sample: Yui)

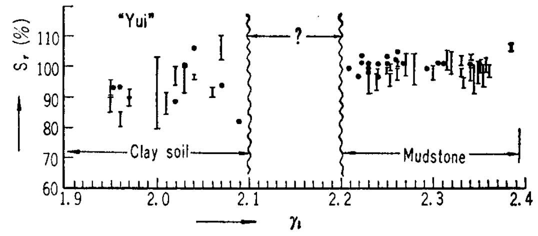

Fig. 9. Relationship between degree of saturation Sr and bulk density γ_t (Sample: Yui)

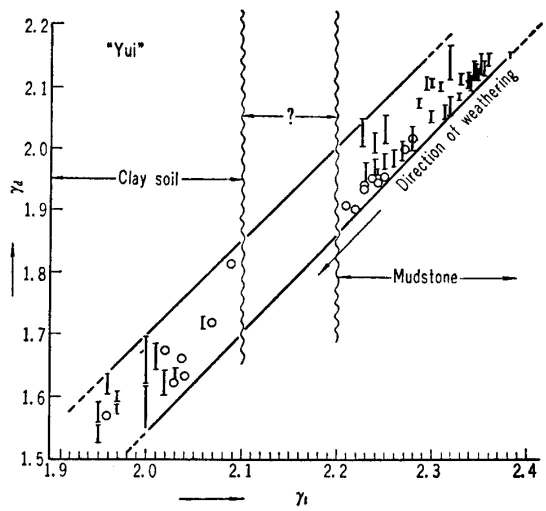

Fig. 10. Relationship between dry bulk density γ_d and bulk density γ_t (sample: Yui)

sical and mechanical properties change accompanying it. It appeares of interest and of practical significance to investigate these changes in detail in realtion to the problems of soundings at landlide areas.

For this purpose unconfined compressive strength q_u, bulk density γ_t, water content W and other physical properties were determined on the mudstones obtained from the bottom of gullies or test pits and those left in water for a long period and on the clay soil.

Some of the Results are shown in Fig. 6~Fig. 10.

The range marked with I in these figures corresponds to the confidence interval of water content from W_1 to W_2.

The " . " shows the case that only one determination of water content was made.

The mark ←?→ shows the gap between clay soil and mudstone. (Such a gap exists between the bulk density of clay soil and that of mudstone which are separated from the mixture of clay soil and weathered mudstone obtained in the test pits.)

Consideration :—

As previously described, there is a sharp difference in the water-resistibility between clay soil and mudstone and, in addition, as Figs. 6–10 show there exists a gap of γ_t between them.

However, looking as a whole, the following relations in physical and mechanical properties exist including clay soil and mudstone.

$$\frac{q_u}{2} = A_1 \exp B_1(\gamma_t - \gamma_0)$$

(From a linear relationship of γ_t and γ_d as shown in Fig. 13,

$$\frac{q_u}{2} = A_2 \exp B_2(\gamma_d - \gamma_{d0})$$

is equivalet to the above).

$$w = A_3 - B_3\gamma_t,\ \gamma_d = A_4 + B_4\gamma_t,\ n = A_5 - B_5\gamma_t\ .$$

$(A_1,\ A_2, \ldots\ldots B_1,\ B_2, \ldots\ldots,\ \gamma_{t_0}$ are constants.)

The same can be affirmed as to the case of Hiraku and Nezumiyashiki. Such a relationship on clay soil has been confirmed many times. It is worthy of notice that similar relationship exists from clay soil through to mudstone.

CONCLUSION

The results are summarized as follows :

1) The weakening of mudstone is due to a kind of chemical dissolution chiefly by hydrogen bonding of originally adsorbed water molecules around clay particles with newly adsorbed ones.

2) It is presumed that a part of Gibbs' free energy evolved by water molecules when adsorbed around clay particles is used as mechanical destructive energy in slaking.

3) In case of Yui and Hiraku, drying in 98% relative humidity is enough to make the mudstones start to slake ; in case of Nezumiyashiki, however, drying in 94% relative humidity is necessary for them to slake.

Among different mudstones there seems to be a difference in the extent of drying necessary to make them start to slake.

4) Mudstone may easily be deteriorated especially within the zone of fluctuating water vapor pressure or fluctuating groundwater level and within geologically fractured zones.

5) There seems to be an intimate relation between water-slaking characteristics of mudstone and landslide occurring on Tertiary sedimentary rocks in Japan.

6) The relations among physical and mechanical properties of mudstone and clay

are expressed as:

$$\frac{q_u}{2} = A_1 \exp B_1(\gamma_t - \gamma_{t_0}) \left(\text{or } \frac{q_u}{2} = A_2 \exp B_2(\gamma_d - \gamma_{d_0}) \right), \quad w = A_3 - B_3 \cdot \gamma_t ,$$

$$\gamma_d = A_4 + B_4 \cdot \gamma_t , \quad n = A_5 - B_5 \cdot \gamma_t$$

ACKNOWLEDGEMENT

The author expresses his deep gratitude to Mr. N. Ohhira and Mr. Y Kayano for their very helpful advice and help.

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