Theory of Time-Temperature-Stress Equivalent Principle Based on Schapery Equation and Its Application on Granite

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The time-temperature-stress equivalent (TTSE) principle refers to the phenomenon that the time-dependent mechanical properties of materials rely on the variations of temperature and stress level. Thus, it is reasonable to predict the long time material mechanical properties based on their relationships with rising temperatures and stress levels. According to the single-integral nonlinear constitutive equation proposed by Schapery, a general expression of the TTSE principle for nonlinear viscoelastic creep property is deduced. The specific expression for time-temperature-stress shift factor is presented by assuming quadratic polynomial form of the Doolittle formula for the viscosity as a function of free volume. Creep curves of granite at different temperatures and stress levels are introduced, and the validation of applying time-temperature-stress principle to predict the long time creep property of granite is provided. The master curves are generated through vertical shift modification and horizontal shift equivalence of all creep curves, and corresponding coefficients in the shift equations are determined by curve fittings. The results indicate the modified time-temperature-stress equivalent relation is appropriate to predict the long time creep property of granite.

Keywords: time-temperature-stress; shift factor; nonlinear creep; granite.

1. INTRODUCTION

The long time performance of mechanical properties of host rock is of great importance in the safety assessment of high-level radioactive waste repository (HLWR), which has service time as long as tens of thousands of years [1, 2]. However, owing to the restrictions of testing conditions, laboratory settings to obtain long time mechanical properties of host rock usually can only reach a time scale of a few days to several months [3, 4]. Obviously, the testable time scale is very short when comparing to the time scale that has to be considered in the long time safety assessment of HLWR. Therefore, the problem of how to predict the long time performance of mechanical properties of rock through short time laboratory data attracts considerable attentions. Based on the studies of the performances of time and temperature change on rock materials [5, 6] and the equivalent relationship between the two variables, it can be inferred that the method employing time-temperature-stress equivalent (TTE) principle is the best choice to solve this problem.

The so-called TTE suggests that the effect of time change on rock mechanical properties is equivalent to the effect of temperature variation. Therefore, it becomes reasonable and convenient to predict the long term performance of mechanical properties of rocks through increasing temperature. The TTE principle is also named time-temperature superposition principle [7] and arises from the studies on viscoelastic property of polymeric materials. According to Leaderman’s [8] observations, the creep compliance vs. logarithmic time curves for different temperatures of various polymeric materials are of the same shape, but the increase of temperature has an effect of contracting the time scale. Tobolsky and Andrews [9] are the first to make use of Leaderman’s observations to superepose the individual curves into a single reference master curve. Ferry [10, 11] presented a theoretical structure for the temperature reduction process in terms of a distribution of discrete relaxation mechanisms. Unlike the wide applications of the TTE principle to polymeric materials [12–15], the studies of the TTE principle on rocks are fewer and just growing in recent years. Paterson [16] and Wang Sheng-zu [17] recommended the application of TTE principle to predict the long time rheological properties for rock-like materials. Liu and his coworkers [18–20] firstly applied the TTE principle to the linear viscoelastic creep property of granite with reliable validation.

However, the linear response range of rock-like materials is usually small when comparing to the total range available prior to yielding or fracturing. The nonlinear property of rock-like materials is often taken into consideration with respect to discussing the long-time stability of rock engineering. Thus, it is necessary to discuss the stress influence on viscoelastic material properties. Based on the fact that high stress can accelerate the creep or relaxation process of material properties, which is similar to the temperature effect, time-stress equivalent (TSE) principle have been proposed for polymeric materials. Brostow [21] studied on the TSE principle from the viewpoint of molecular chain. Yen [22] investigated the accelerated characterization of the creep response of a unidirectional off-axis composite using TTSE principle. Luo and co-workers [23, 24] proposed the characterization of TTSE principle in terms of the relation
between viscosity and free volume and corresponding shifting method to generate the master curves is presented.

The objectives of this study are to build the general relationship for the temperature-stress principle based on Schapery’s single-integral nonlinear constitutive equation, and to present an appropriate equation with addition of horizontal and vertical shift factors to describe the equivalent relation, and conduct the validation of applying TTSE principles to the nonlinear creep property of granite.

2. THEORY OF THE TTSE PRINCIPLE

2.1. Characterization of the TTSE principle

The thermodynamic theory developed by Schapery permits us to express materials properties in terms of either stress or strain. When stress is treated as an independent state variable, the theory yields the following equation [25]:

\[ \varepsilon = g_0 J_0 \sigma + g_1 \int_0^t \Delta J (\varepsilon - \psi') \frac{dg_2 \sigma}{d\tau} d\tau, \]  

where \( J_0 \) and \( \Delta J \) are the components of the linear viscoelastic creep compliance, \( \varepsilon \) is called reduced-time defined by

\[ \psi = \psi(t) = \int_0^t \frac{d\tau'}{a_0 (\sigma(\tau'))} \]  

and

\[ \psi' = \phi(z) = \int_0^t \frac{d\tau'}{a_2 (\sigma(\tau'))} \]  

(1)

(2)

(3)

where \( g_0, g_1, \) and \( a_2 \) are functions of stress which represent the nonlinear material properties. These stress-dependent properties have specific thermodynamic significance. The changes of \( g_0, g_1, \) and \( g_2 \) reflect the third or higher order of dependence of the Gibbs free energy on the applied stress, and \( a_0 \) arises from similar high-order effects in both entropy production and free energy.

Substituting a constant stress in Eq. (1), and using Eq. (2), the nonlinear creep compliance \( J_n \) is obtained as follows

\[ J_n = \frac{\varepsilon}{\sigma} = g_0 J_0 + g_1 g_2 \Delta J (\frac{t}{a_0}). \]  

(4)

There are a considerable amount of published creep curves on a variety of materials which fit the special cases expressed in Eq. (4). Obviously, Eq. (4) can be simplified to the form of modified superposition principle when \( g_0 = g_1 = g_2 = a_0 = 1 \). Therefore, this nonlinear equation is just developed by introducing nonlinear coefficients to linear viscoelastic constitutive equation from a mathematical viewpoint.

For convenience of subsequent illustration, the relation describing the linear viscoelastic TTE principle can be simplified as follows

\[ J(T_n, t) = J(T_n, t / a_T). \]  

(5)

where \( T_n \) is the reference temperature and \( T \) is an arbitrary temperature; \( a_T \) is a horizontal shift factor, representing equivalent relationship between changes of temperature and variations of time scale.

According to different time dependencies, strain can be put in the following form

\[ \varepsilon = \sigma / J_0 + \sigma / \Delta J(t), \]  

where \( J_0 \) is the initial value of compliance and \( \Delta J(t) = J(t) - J_0 \) is the transient component of compliance.

With Eq. (5) and Eq. (6), the relations in the TTE principle can be rewritten as follows

\[ J_n(T_n) = J(T); \]  

\[ \Delta J(T_n, t) = \Delta J(T, t / a_T). \]  

(7)

(8)

Similarly to the TTE principle, the reference stress \( \sigma_r \) and an arbitrary stress \( \sigma \) are defined firstly when it comes to the TTSE principle.

According to Eq. (4), the nonlinear viscoelastic creep is composed of two components. One is time-independent initial creep component \( g_0 J_0 \), the other is highly time-dependent transient \( \Delta J_n \). Obviously, it is easy to obtain the stress dependence of initial creep using creep tests at different stresses.

From Eq. (4), the nonlinear transient creep is written as

\[ \Delta J_n = J_n - g_0 J_0 = g_1 g_2 \Delta J (\frac{t}{a_0}). \]  

(9)

Substituting \( a_0 \) with two stress states \( \sigma_r \) and \( \sigma \) in Eq. (9), the following equations are obtained.

\[ \Delta J_n (\sigma, T, t) = g_1 (\sigma) g_2 (\sigma) \Delta J(T, \frac{t}{a_0 (\sigma)}); \]  

\[ \Delta J_n (\sigma_r, T, t) = g_1 (\sigma_r) g_2 (\sigma_r) \Delta J(T, \frac{t}{a_0 (\sigma_r)}). \]  

(10)

(11)

The relation in Eq. (10) can be put in the equivalent form:

\[ \Delta J(T, \frac{t}{a_0 (\sigma)}) = \Delta J(T, t / a_T (T, \sigma)). \]  

(12)

Combining Eq. (12) to Eq. (10), the relation can be written as:

\[ \Delta J_n (\sigma, T, t) = g_1 (\sigma) g_2 (\sigma) \Delta J(T, \frac{t}{a_T (T, \sigma)}). \]  

(13)

Define

\[ t' = \frac{a_T (T, \sigma)}{a_0 (T, \sigma)} t. \]  

(14)

Taking Eq. (14) into Eq. (13) and replacing \( t' \) with \( t \), the following equation is obtained

\[ \Delta J_n (\sigma, T, t) = g_1 (\sigma) g_2 (\sigma) \Delta J(T, \frac{t}{a_T (T, \sigma)}). \]  

(15)

The logarithmic forms of Eq. (11) and Eq. (15) are:

\[ \lg \Delta J_n (\sigma, T, t) = \lg g_1 (\sigma) g_2 (\sigma) + \lg \Delta J(T, \frac{t}{a_T (T, \sigma)}); \]  

\[ \lg \Delta J_n (\sigma, T, t) = \frac{a_T (T, \sigma)}{a_0 (T, \sigma)} = \frac{a_0 (T, \sigma)}{a_T (T, \sigma)} \]  

\[ = \frac{a_T (T, \sigma)}{a_0 (T, \sigma)} \frac{\lg g_1 (\sigma) g_2 (\sigma) + \lg \Delta J(T, \frac{t}{a_T (T, \sigma)})}{\lg g_1 (\sigma) g_2 (\sigma) + \lg \Delta J(T, \frac{t}{a_T (T, \sigma)})}. \]  

(16)

(17)

Using Eq. (16) to minus Eq. (17), the following equation is obtained

\[ \lg \Delta J_n (\sigma, T, t) = \frac{\lg g_1 (\sigma) g_2 (\sigma)}{a_T (T, \sigma)} + \frac{a_T (T, \sigma)}{a_0 (T, \sigma)} \]  

\[ + \frac{a_0 (T, \sigma)}{a_T (T, \sigma)} \frac{\lg g_1 (\sigma) g_2 (\sigma) - \lg g_1 (\sigma) g_2 (\sigma)}{\lg g_1 (\sigma) g_2 (\sigma) - \lg g_1 (\sigma) g_2 (\sigma)} \]  

It is reasonable to define

\[ a_0 (T, \sigma) = g_0 (\sigma) = g_1 (\sigma) = g_2 (\sigma) = 1, \]  

then Eq. 18 becomes

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\[
\log \Delta J_n(\sigma_r, T_r, t) = \\
= \log \Delta J_n(\sigma, T, t) - \frac{a_T(\sigma)}{a_T} - \log g_1(\sigma)g_2(\sigma).
\] (20)

Eq. (20) represents the nonlinear viscoelastic TTE principle of transient compliance. It can be seen that the stress and the transient compliance \( \Delta J_n \) at the reference temperature can be expressed using a random temperature and stress. Therefore, the nonlinear TTE principle is also regarded as time-temperature-stress equivalent principle.

### 2.2. Shift factors

According to the equivalent theory \([11]\), the horizontal shift factor is defined as follows:

\[
\log a = \frac{\log \tau}{\log \eta} = \frac{\log \eta - \log \eta_0}{\log \eta},
\] (21)

where \( \tau \) and \( \eta_0 \) represent the relaxation time at a random state and a reference state, respectively; \( \eta \) and \( \eta_0 \) are the viscosity coefficient at a random state and a reference state, respectively.

Based on the free volume theory \([11]\), the viscosity of the material is related to the free volume fraction by Doolittle equation

\[
\log \eta = A + B(\nu - \nu_f)/\nu_f,
\] (22)

where \( A, B \) are material constants, \( \nu_f \) is free volume, \( \nu \) is total volume.

For polymeric materials, such as rayon, nylon, and cellulose, the variation of free volume with temperature and stress is usually assumed to be linear \([11]\). However, considering the complexity of mechanical properties for rock-like materials, it is reasonable to assume satisfying the following quadratic polynomial form

\[
f = f_0 + \alpha_T(T - T_0) + \beta_T(T - T_0)^2 + \alpha_\sigma(\sigma - \sigma_0) + \beta_\sigma(\sigma - \sigma_0)^2,
\] (23)

where \( f = \nu_f/\nu \) as the fractional free volume; \( \alpha_T \) and \( \beta_T \) are thermal expansion coefficients of the fractional free volume; \( \alpha_\sigma \) and \( \beta_\sigma \) are stress-induced thermal expansion coefficients of the fractional free volume; and \( f_0 \) is the fractional free volume at the reference state.

Substituting Eq. (22) and Eq. (23) into Eq. (21), the horizontal shift factor is obtained

\[
\log a_c = C_1 \left( C_2(T - T_0) + C_3(T - T_0)^2 + C_4(\sigma - \sigma_0) + C_5(\sigma - \sigma_0)^2 \right) \frac{C_2 + C_3(T - T_0) + C_4(T - T_0)^2}{C_2 + C_3(T - T_0) + C_4(T - T_0)^2}.
\] (24)

where we define

\[
C_1 = \frac{R}{f_0}, \quad C_2 = f_0, \quad C_3 = \alpha_T, \quad C_4 = \beta_T, \quad C_5 = \alpha_\sigma, \quad C_6 = \beta_\sigma.
\] (25)

In addition, we define the temperature shift factor at a constant stress level and the stress shift factor at a constant temperature in such a way that

\[
\eta(T, \sigma) = \eta(T_0, \sigma)\alpha_T(\sigma) = \eta(T_0, \sigma_0)\alpha_\sigma(\sigma_0)
\] (26)

Comparing Eq. (21) with Eq. (26), it is found that the combination shift factor is expressed by temperature shift factor and stress shift factor as follows

\[
a = a_T\sigma_0 \alpha_T = a_\sigma(\sigma_0)\alpha_\sigma \sigma.
\] (27)

It is seen from Eq. (26) and Eq. (27) that the viscoelastic material properties can be shifted along the time scale to generate a master curve of a longer time scale at a reference temperature \( T_0 \) and a reference stress \( \sigma_0 \) within one step if using the temperature-stress shift factor \( a \) or in two steps if using a combination of stress shift factor \( a_T \) and temperature shift factor \( a_\sigma \).

When temperature keeps consistent at the reference temperature, the stress shift factor can be obtained from Eq. (24) as follows

\[
\log a_T = C_1 \frac{C_1(T - T_0) + C_2(T - T_0)^2}{C_2 + C_3(T - T_0) + C_4(T - T_0)^2}.
\] (29)

Similarly, if stress keeps invariant at the reference stress, the temperature shift factor can be obtained from Eq. (24) as follows

\[
\log a_\sigma = C_1 \left( C_2(T - T_0) + C_3(T - T_0)^2 + C_4(\sigma - \sigma_0) + C_5(\sigma - \sigma_0)^2 \right) \frac{C_2 + C_3(T - T_0) + C_4(T - T_0)^2}{C_2 + C_3(T - T_0) + C_4(T - T_0)^2}.
\] (31)

In contrast to the WLF equation \([10]\) for the linear viscoelastic properties, the Eq. (29) can be seen as a modified form of the WLF equation.

With the stress shift factor, the nonlinear creep behavior can be characterized by the stress-induced equivalent time

\[
J(\sigma, t) = b_\sigma \cdot J(\sigma_0, t / a_\sigma).
\] (30)

where \( b_\sigma \) is the vertical shift factor which reflects the stress influence on the material microstructure. It is assumed to be the same form to the horizontal shift factor

\[
\log b_\sigma = V_1 \cdot V_3^\sigma(\sigma - \sigma_0) + V_2^\sigma(\sigma - \sigma_0)^2.
\] (31)

where \( V_1, V_2, V_3^\sigma \) and \( V_4^\sigma \) are the coefficients. It is determined through experimental tests.

### 2.3. Procedures

Before discussing the procedures for the TTSE principle, for convenience of illustration, the procedures for the TTE principle is firstly introduced. According to Eq. (5), the TTE principle has been widely applied to obtain the master curves for different rheological properties, such as creep strain, creep compliance, and stress compliance against logarithmic time or dynamic modulus against frequency. In order to generate a creep compliance master curve, the following steps are required, and the corresponding graphical representation is shown in Fig. 1.

![Fig. 1. Procedures and criteria for applying the TTE principle](image)

a – Conventional compressive creep tests of specimens are performed at different temperatures under a constant load, and the variation of creep compliance curves are plotted against logarithmic time.
b – A reference temperature somewhere within the range of the testing temperatures is arbitrarily selected.

c – All the individual creep compliance curves except the one at the reference temperature are shifted along the logarithmic time axis to compose a master curve.

Ferry [10] commented that the TTE principle can be established when (1) the exact matching of shapes of adjacent curves is obtained; (2) the same value for all viscoelastic functions; and (3) the temperature dependence has a reasonable form such as the Arrhenius or the WLF equations. If the TTE principle is valid, the master curve represents the real behavior of a long time test at the reference temperature. In the absence of actual long time test data, mathematical optimization methods are always performed to minimize the discrepancy between the adjacent curves when they are overlapped. This actually turns the necessary condition into a sufficient condition to allow the reliance on the TTE principle.

If it is possible to generate a smooth master curve by applying a horizontal shift along the logarithmic time axis, the material is classified as a thermorheologically simple material. However, for some materials a vertical shift factor may be additionally needed to obtain a smooth master curve, so in this case they are classified as thermorheologically complex materials [9].

Comparing Eq. (5) with Eq. (20), it can be found that they are essentially of the same expression, which indicates that the procedures are similar. However, the vertical shift factor in the TTE principle is frequently ignored, while often included in the TSE principle. Thus, before the horizontal shift is performed, that is before procedure (c), the creep curves have to be shifted vertically for achieving the TSE principle.

3. EXPERIMENTAL RESULTS

The creep test data of granite at different temperatures and stresses have been published by Rummel [26] are adopted in this paper to do the validation of applying time-stress equivalent principle to the creep property of granite, since the long time performance of mechanical properties of granite plays an important role in the safety assessment of high-level radioactive waste disposal.

The granite blocks for the test are from the quarry near Flossenburg and machined to the specimens with a diameter of 3 cm and a height of 12 cm. The grain sizes are between medium and coarse, and the volume percentages of the minerals are 20 % for quartz, 30 % for microcline, 12 % for white mica, 2 % for black mica, and 1 % for other components.

Uniaxial creep tests are performed by the 60 t press machine with loading stress levels of 40 MPa, 64 MPa, and 80 MPa and heating temperatures of 20 °C, 100 °C, 150 °C, 200 °C, 400 °C respectively. The loading pressure is monitored by the spring pressure gauge. The axial and hoop strain are measured by resistance strain gauge. To ensure free deformation of the samples, the lubricant MoS₂ is applied to the granite specimens. The testing time for each specimen is approximately 6×10³ seconds.

As shown in Fig. 2 and Fig. 3, the creep compliance \( J \) curves and creep rate curves at different temperatures and stress levels are presented. From these curves, two stages can be seen in the whole creep process: one is the decelerating creep stage, and the other is a steady-state creep stage. In the decelerating creep stage, the creep rates at different stresses are closely distributed, which indicates that the adjustment of mineral fabric presents a random property at the early stage. However, the creep rate apparently increases with rising of stress levels at the same
temperature until the creep process turns into a steady state. This is similar to the temperature effect on creep property of granite.

**4. APPLICATIONS OF THE TTSE PRINCIPLE**

The TTSE principle can be simplified respectively to the TSE and TTE principle when temperature and stress keep consistent. The validity of TTE principle has been approved. Below only presents the application of the TSE principle to the nonlinear creep property of granite.

The reference stress chosen for this study is the lowest stress 40 MPa. According to the procedures of the TSE principle, the master curves at different temperatures finally are generated (Fig. 4) by two steps. Firstly, the curves are shifted by eye until a smooth match is obtained. Then, this is subsequently refined by a computer-based optimization method in which fine adjustments are made to the shifts. The specific procedures are (1) Select data points for optimization. The overlapping part between the adjacent creep curves are selected as the data points since the smoothness of the master curve only determined by this region. (2) Choose an appropriate polynomial function to fit the data points. Data points are fitted to a 2nd order polynomial in this paper. (3) Calculate the optimal value of \( \log(a_T) \). The undetermined value of the shift factor \( \log(a_T) \) is altered numerically to minimize the sum of square error between the data points and the 2nd order polynomial function. The corresponding vertical and horizontal shift
values are obtained in Table 1 and Table 2, respectively. The master curves show good smoothness, which indicates the validity of applying time-stress principle to predict long-time creep property of granite. The time scales of the creep property in the master curves range from $3.6 \times 10^3$ s to $6.0 \times 10^5$ s, $6.0 \times 10^5$ s, $6.0 \times 10^5$ s, and $7.5 \times 10^5$ s when temperatures are 20°C, 100°C, 150°C, 200°C and 400°C, respectively.

Fitting the data in Table 1 and Table 2 with Eq. (28) and Eq. (31), the coefficients can be determined by the LM (Levenberg-Marquardt) optimization algorithm (Fig. 5; Fig. 6). The specific forms for vertical shift factor and horizontal shift factor are shown in Eq. (32) and Eq. (33), respectively. The square correlation coefficient $R^2$ of vertical shift factor in Eq. (33) is relatively small. This indicates the discreteness of the vertical shift results, caused by the heterogeneity of the granite samples. The correlation between the experimental results and the theoretical results is good, which reflects that the quadratic polynomial form presented in this paper is useful to describe the equivalent relation between time and stress for the creep property of granite.

Table 1. Shift values of $\log b_\sigma$ at different stresses

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Stress level (MPa)</th>
<th>40</th>
<th>64</th>
<th>87</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>0</td>
<td>−0.05</td>
<td>−0.10</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>0</td>
<td>−0.10</td>
<td>−0.15</td>
<td></td>
</tr>
<tr>
<td>150</td>
<td>0</td>
<td>−0.08</td>
<td>−0.25</td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>0</td>
<td>−0.08</td>
<td>−0.30</td>
<td></td>
</tr>
<tr>
<td>400</td>
<td>0</td>
<td>−0.06</td>
<td>−0.28</td>
<td></td>
</tr>
</tbody>
</table>

Table 2. Shift values of $\log a_\sigma$ at different stress levels

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Stress level (MPa)</th>
<th>40</th>
<th>64</th>
<th>87</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>0</td>
<td>−0.50</td>
<td>−1.25</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>0</td>
<td>−0.45</td>
<td>−1.12</td>
<td></td>
</tr>
<tr>
<td>150</td>
<td>0</td>
<td>−0.55</td>
<td>−1.05</td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>0</td>
<td>−0.56</td>
<td>−1.06</td>
<td></td>
</tr>
<tr>
<td>400</td>
<td>0</td>
<td>−0.58</td>
<td>−1.02</td>
<td></td>
</tr>
</tbody>
</table>

It should be pointed out that the master curves eventually generated in Fig. 4 indicate accelerated description of approximately 10 times beyond the test duration. Therefore, it is effective to employ the TTSE principle to predict the long-time creep property of granite. However, the time scale of the master curves obtained in this paper is still not adequate to assess the long time safety of the high level radioactive waste repository. Higher temperatures and stress levels creep tests should be performed if we want to obtain the longer-time testing data.

\[
\log b_\sigma = -0.677 \left( \frac{(\sigma - 40) + 4.587(\sigma - 40)^2}{21720 + (\sigma - 40) + 4.587(\sigma - 40)^2} \right) \quad (33)
\]

\[
(R^2 = 0.8900)
\]

6. CONCLUSIONS

From the results obtained in the present study, the following conclusions can be drawn:
1) The TTSE principle can be used in the prediction of long-time creep property of granite.
2) Both vertical stress shift factor and horizontal stress shift factor have to be considered in generating master curves.
3) It is reasonable to assume the fractional free volume satisfying the quadratic polynomial form for rock-like materials. That is to say, the formula of the vertical and horizontal stress shift factor presented in this study can be applied to the long time prediction of creep property for rock-like materials.

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