

Short Commentary

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Modeling of Nanodroplets and Nanocylinders

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Abstract

In this review we consider formula that determines the curvature dependent surface tension in a droplet with two phases. Taking into account the size dependence of the surface tension systems of nonlinear differential equations describing the droplet profile are obtained. We have shown if the droplet size is not so large compared to the thickness of the surface layer (micro- or nanodroplets) the dependence of the surface tension on the curvature is very important.

Keywords: Droplet Profile; Gibbs Adsorption; Nanodroplet Models; Tolman Length; Two-dimensional Heisenberg model twodimensional

Introduction

The nonwetting liquid takes the form of a drop on a surface of a body. The well-known Young equation describes the balance at the threephase contact of solid-liquid and gas. The interfacial tensions form the equilibrium contact angle of wetting, many times referred to as Young contact angle, θ . Young suggested measuring contact angle θ of the liquid with the surface. The mechanical equilibrium of a droplet lying on a flat hard surface under the constraints of three surface tensions is called the Young equation. The surface tension (Young's equation) for large droplets is determined **[1-10]** (see figure 1) by

$$\sigma_1 = \sigma_2 + \sigma \cos \theta$$
, (1a)

where σ_1 is tension at the interface of the solid and liquid phases, σ_2 is tension at the interface of the solid and vapor phase, σ is tension at the interface of the liquid and vapor phases. The equilibrium contact angle reflects the relative strength of the liquid, solid, and vapours molecular interaction.

The drop volume, V, is determined from angle θ from the formula [1-9]:

 $V \sim \frac{1}{3}\pi r_s^3 \phi(\theta), \quad (\mathbf{1b})$

where r_s is equilibrium radius of a drop, and

$$\phi(\theta) \sim (1 - \cos \theta)^2 (2 + \cos \theta)$$
. (1c)



Figure1: The dimensionless profiles of sessile microdrop.

Let us consider an isolated system in thermodynamic equilibrium, which consists of two bulk phases with different densities - steam and liquid and also the interface between them.

Here the surface tension acts as a separating surface with a *minimum value* [1-3]. Excess pressure is provided by the Laplace equation [4-10]:

$$\Delta p = \kappa \sigma \, (2a)$$

where σ is the surface tension, κ determines the mean curvature of the surface. Differential of excess pressure

$$d(\Delta p) = \kappa \, d\sigma + \sigma \, d\kappa \,. \tag{2b}$$

To determine the surface tension, let us apply the Gibbs equation that can be written in the form of

$$d\sigma = -\Gamma d\mu = -\delta d(\Delta p), \qquad (3)$$

where Γ is the Gibbs adsorption, μ is the chemical potential, $\delta \ge 0$ is a non-negative parameter characterizing the thickness of the interfacial layer. In surface thermodynamics the Tolman length is used as a parameter δ which is equal to the distance between the surface of tension and equimolar surface. The numerical values of parameter δ are in the range from 0.1 to 1 nm far from the critical boiling point. When writing the equation (3) we have taken into account that the differential for the chemical potential and the Gibbs adsorption are equal $d\mu = d(\Delta p)/\Delta n$ and $\Gamma = \delta \Delta n$, where Δn is the substance concentrations difference in interfacial phases. Equation (3) holds for any smooth interfacial phase irrespective of its geometrical shape. In what follows we assume that δ does not depend on the curvature radii. This assumption is considered acceptable if the curvature of the surface is not too large compared with $1/\delta$ [1,2]; an analysis of the size dependence of the surface tension for the spherical surface show that such assumption is already acceptable when $|\kappa| < 0.1/\delta$. Substituting (2) into (3) we can obtain the equation

$$\frac{d\sigma}{\sigma} = -\frac{\delta}{1+\delta\kappa}d\kappa \tag{4}$$

Having integrated (4) we can find

$$\sigma = \frac{\sigma^{(\infty)}}{1 + \delta \kappa},$$
(5)

where $\sigma^{(\infty)}$ is a flat surface tension as $\kappa \to 0$. For arbitrarily curved surface $\kappa = 1/r_1 + 1/r_2$, where r_1 and r_2 are the principal radii of curvature of the surface [9] therefore from (5) we finally obtain

$$\sigma = \frac{\sigma^{(\infty)}}{1 + \delta\left(\frac{1}{r_1} + \frac{1}{r_2}\right)}.$$
(6)

As can be expected, when $r_1 = r_2$ from (6) the well-known Tolman formula is derived for small spherical droplets [3]. When $|r_1| >> |r_2|$ we have a formula for a long and thin-walled cylinder of radius r_2 [4]. We also note that the above arguments are in accordance with the provision that at sufficiently high r_1 and r_2 in the thermodynamic equations for the spherical surface curvature can be replaced by medium (Euler) curvature.

In [9] a comparison of simple size dependency for spherical and cylindrical shaped surfaces resulted in the following interpolation formula

$$\sigma = \sigma^{(\infty)} \left[1 - \delta \left(\frac{1}{r_1} + \frac{1}{r_2} \right) \right].$$
(7)

It is easy to verify that formula (7) can be derived using expansion of (5) into series of $\delta \kappa$ in view of zero and consider first member only.

Theoretical remarks for Rekhviashvili - Sokurov's model

As the most important application of the above results from a practical point of view, let us consider the problem of the sessile droplet. The droplet is on the poorly wet table substrate for example on the graphite atomically smooth surface.



Figure 2: The sessile drop profile corresponding to surface of tension.

The dotted line is the equimolar surface with the zero Gibbs adsorption ($\Gamma = 0$).

We use the Bashforth-Adams approximation (see [6-9]) and according to this, the influence of the substrate is not taken into account. From the standpoint of thermodynamics, the droplet is macroscopic object. The solid-liquid interface is flat, so the size dependence of surface tension is not considered here. Origin is superimposed with droplet vertex which is on the surface of tension. The equilibrium droplet profile is determined in terms of the constancy of the sum of hydrostatic and capillary pressure. In this case the Laplace equation is

$$\sigma\left(\frac{1}{r_1} + \frac{1}{r_2}\right) = p_0 + \Delta\rho g z , \qquad (8)$$

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$$\frac{1}{r_1} = \frac{d\varphi}{ds}, \quad \frac{1}{r_2} = \frac{\sin\varphi}{x}, \quad \varphi \in [0, \pi],$$

where P_0 is the pressure of the droplet measured at the top (x = 0, z = 0), φ is slope of the tangent at a point of the meridian, *s* is the arc length, *x* and *z* are the coordinates that define the droplet cross-section, $\Delta \rho = m \Delta n$ is the difference in density between the liquid and gaseous phases, g is the gravitational acceleration, *m* is the mass of a single particle (atom or molecule). In this case the surface tension depends on the local curvature of radii r_1 and r_2 . Substituting (6) into (8) we obtain

$$\frac{d\varphi}{ds} + \frac{\sin\varphi}{x} = \frac{2\sigma^{(\infty)} + \Delta\rho g z(r+2\delta)}{\sigma^{(\infty)}(r+2\delta) - \delta[2\sigma^{(\infty)} + \Delta\rho g z(r+2\delta)]},$$
(9)

$$\frac{dx}{ds} = \cos\varphi, \quad \frac{dz}{ds} = \sin\varphi, \quad (10)$$

$$x(s=0) = z(s=0) = \varphi(s=0) = 0,$$
 (11)

where *r* is the radius of curvature of the droplet vertices. When deriving equation (9) we took into account that because of the axial symmetry of the droplet on its top the following condition is hold [9]: $r = r_1 = r_2 = 2\sigma^{(\infty)} / p_0$. As $\delta = 0$ the surface tension dependence on the curvature is absent. In this case, it follows from (9) exactly the well-known Bashforth-Adams equation [9, 10]. The Bashforth-Adams equation is reduced to various differential equations for the droplet profile or volume which are solvable numerically.

If we introduce the dimensionless (related to r) variables x and z, then (9) - (11) are reduced to the Cauchy problem for a system of two nonlinear differential equations

$$\frac{dx}{d\varphi} = \frac{[1 - \alpha(\gamma + \beta z)]x\cos\varphi}{(\gamma + \beta z)x - [1 - \alpha(\gamma + \beta z)]\sin\varphi},$$

$$\frac{dz}{d\varphi} = \frac{[1 - \alpha(\gamma + \beta z)]x\sin\varphi}{(\gamma + \beta z)x - [1 - \alpha(\gamma + \beta z)]\sin\varphi},$$
(13)

$$\frac{dV}{ds} = \pi x^2 \sin \phi , \qquad (14)$$

Or

$$\frac{dV}{d\phi} = \pi \frac{\left[1 - \alpha \left(\gamma + \beta z\right)\right] x^3 \sin \phi}{\left(\gamma + \beta z\right) x - \left[1 - \alpha \left(\gamma + \beta z\right)\right] \sin \phi}$$

$$x(\varphi = 0) = z(\varphi = 0) = 0$$
, (14a)

 $\alpha \ge 0$, $\beta > 0$, $0 < \gamma \le 2$,

where $\alpha = \delta / r$, $\beta = \Delta \rho g r^2 / \sigma^{(\infty)}$ and $\gamma = 2/(1+2\alpha)$ are dimensionless parameters that define the physical properties for the droplet. These equations contain only two independent parameters α and β which depends on the specific experimental

conditions: the parameter α determines the dependence of the surface tension on the surface curvature; the parameter β is linked to the capillary constant $c = r^2 / \beta$.

Equations (12), (13) with the initial conditions (14) were solved numerically by the 4th order Runge-Kutta method with fixed step size. Graphs of the resulting solutions are shown in **Figure 2a,b**. For clarity in these figures the origin is shifted to the droplets bottom. The algorithm has been verified at $\alpha = 0$ using the Bashforth-Adams table data which is partially presented in [9]. This way reveals that the coincidence of the solutions is ensured at four significant digits. Since error in experimental measurements within the sessile drop method is, as a rule, higher then 0.1% then the accuracy achieved can be considered satisfactory for the solution of the equations.

Figure 3a. shows that with the increase of the parameter β the droplet on the substrate surface becomes flatter. It may be due to a decrease in surface tension $\sigma^{(\infty)}$ and/or increase in the droplet density. Here the size effect of surface tension is not considered that corresponds to the parameter values: $\alpha = 0$, $\sigma = \sigma^{(\infty)}$ and $\gamma = 2$.

But it follows from the above models under certain conditions the dependence of the surface tension of the radii of curvature affects the equilibrium shape of the droplet and it is shown in **Figure 3b**. With the increase of the parameter α the droplet flattened. This is due to a general decrease of surface tension and increase of the interface layer thickness. The change in the profile, in view of the above reasons, is not visible for larger drops. However, for micro- and nanosized droplets this could play a significant part at higher temperatures. It should be noted that the nanodroplets being a common modern nanotechnology objects are studied with transmission electron microscopy that provides high-resolution for studding the droplet shape.



Figure 3 a, b: The dimensionless profiles of sessile drops at various values of the parameters α and β [9]. If the droplet profile is known, it is possible to calculate the droplets equilibrium volume. In general, it is equal to

$$V = \pi r^3 \int_{0}^{z_0} x^2 dz$$
, (15)

where z_0 is the *dimensionless meniscus height* defined as a maximum value in the coordinate system. Our calculations showthat the sessile droplet volume can be well approximated by the following empirical formulas:

$$V \sim \frac{5 r^{3}}{\beta + 1} \exp(-2, 5\beta^{0.4} \alpha)$$
, (16a)
$$V \sim \frac{9r^{3}\beta^{-2}}{\alpha + 0, 2\beta^{-1} + 0, 2}$$
. (16b)

For small values of the parameter ($\beta \sim 0.1$) the calculated error in the sessile droplet volume according to formula (16a) does not exceed 5%. Increase in β parameter leads to this error rapid decrease. Size effect of the surface tension can be neglected when $\alpha \rightarrow 0$ and formula (16a) remains valid. We use formula (16b), if $\beta > 1$.

Computational experiment for Rekhviashvili - Sokurov's model

Let us discuss briefly the possible application of the system of equations (12), (13). To determine the surface tension by the sessile drop method taking into account the curvature dependence of surface tension we can use an independent assessment of the surface layer thickness and the radius of curvature at the droplet apex obtained for example by using transmission electron microscopy.

We show the calculated nanodroplet models.



Figure 4a: Model image of sessile nanodrop ($\alpha = 0.2$, $\beta = 10$) [9].





Figure 4c: Model of sessile nanodrop at $\alpha = 0:1$, $\beta = 5$ [10].

The thickness of the surface layer is taken as the maximum value of δ [1]. Once the parameters δ and r are determined the numerical solution of equations (12), (13) is required as well as its correlation with the sessile droplet profile to determine the parameter of β .

This method is easier to implement but, generally speaking, it requires mathematical foundation: by correlation of the droplet profile and numerical solution of equations (12), (13) two parameters of α and β are extricated by this or that method.

We will develop new algorithms, and also computing experiments.

Methods for detailed determining of the surface tension without considering size effects are studied in [5-7, 10]. For the above approximation these techniques are quite constructive.

The next step in theoretical studies will be modification of the equations (12), (13) taking into account Van der Waals forces between the droplet and the substrate.

The Long Nanocylinder

We consider a case of the application theory [8, 9], when nanoparticles has a long cylinder form. We used the cylindrical coordinate system for which the characteristic spin function [8, 9] is presented by the angle function q(r) about the cylinder axis z.

$$H_{g,c} = \frac{A}{2} \left[\theta'^2 + \frac{\sin^2 \theta}{r^2} \right],$$
(17)

The free energy in this model has the following form [8, 9]:

where $\theta(r)$ is the angle between the cylinder axis and the magnetization vector; r is the radial coordinate.

The model kinetic energy in (17) is a classical analog of the exchange energy in the Heisenberg model for the two-dimensional space at the continuum approximation **[8]** that in our case corresponds to the infinite cylinder model **[8, 9]**. Then the kinetic energy in (17) coincides in form with the kinetic energy of particle (in cylindrical coordinates). This fact is not casual as the model under consideration permits exact analytical solutions in the form of quasiparticles (nonlinear waves) which are called instantons (or skyrmions **[8, 9]**). Please note that in our case these quasi--particles are not dynamic particles but topological compositions. Therefore, in our case the virtual kinetic energy of the topological instanton is meant by the kinetic energy.

We introduce a relative coordinate:

$$\rho = \frac{r}{R_c},$$
 (18)

where R_c is the drop equilibrium radius.

Then there is a condition of $0 \le \rho \le 1$. The proposed continuum model of energy (17) actually appears a Heisenberg model in which the interacting spins have meaning of the energy states of the particles associated with the constant exchange interaction *A* (with the dimension for the exchange energy [J/m]).

Using (17) it is simple to derive the Euler - Lagrange equation:

$$\theta''(\rho) + \frac{\theta'(\rho)}{\rho} - \frac{\sin\theta\cos\theta}{\rho^2} = 0.$$
 (19)

For simplicity it is sufficient to use only a particular solution of this equation describing the nucleation process under simple boundary conditions:

$$\theta(\rho) = \begin{cases} \pi, \ \rho = 0, \\ \frac{\pi}{2}, \ \rho = 1. \end{cases}$$
(20)

The solution of problems (19), (20) has a simple form:

$$\tan\left(\frac{\theta}{2}\right) = \frac{1}{\rho},$$
 (21)

that is convenient for further analysis.

Let us introduce the model surface energy in order to obtain the Euler - Lagrange equations for the scale - invariant theory as well:

$$\theta_a''(\rho) + \frac{\theta_a'(\rho)}{\rho} - \frac{a^2 \sin \theta_a \cos \theta_a}{\rho^2} = 0,$$
(22)

where a^2 is the ratio of the anisotropy energy to the exchange interaction constant A. The parameter a^2 is determined in [8, 9]:

$$a^2 = \frac{B}{A} + 1$$
, (23)

with the determined anisotropy function (model as the Rapini modified potential (see [8, 9])):

$$\frac{B\sin^2\theta_a}{2\rho^2},$$

where *B* is the positive energy quantity whose dimension coincides with *A*.

For agreement with the previous solution we assume that in (22) at B = 0 there is no anisotropy, and at B > 0 it occurs. The solution of equation (22) is as follows:

$$\tan\left(\frac{\theta_a}{2}\right) = \frac{1}{\rho^a}.$$
 (25)

Please note that solutions (21) and (25) analytically join, therefore the indices are later omitted.

Let us consider one general solution (25). The diagram of this solution is presented in Figures 1a, b.

It is easy to show that the function $\theta(r)$ at a = 1 and $0 \le \rho \le 1$ has no point of inflection. This point appears only at a > 1. This means that the surface layer in our model can exist only at a > 1. In this case some volume whose energy being the surface energy of the cylindrical particle can be chosen as a surface layer. For definiteness we suppose, for example, that the surface layer begins to manifest itself clearly from a value of a > 4. Thus, we suppose that if a = 1 there is no anisotropy in the system, and the Tolman length actually coincides with the drop sizes. If a >> 1, then in the proposed model the specific anisotropy is more than the exchange interaction, and in the drop there appears a parameter (the Tolman length) which characterizes the dimension of the interfacial region.



Figure 5a: Diagrams of solution (25) at different values of parameter a (see [9]).



Figure 5b: Diagrams of solution (25) at different values of parameter a:

1 - *a*=1; 2 - *a*=10; 3 - *a*=50; 4 - *a*=100. (See **[8]**)

The case a < 1 corresponds to the negative surface energy (in **Figure 5a** this case is presented for a = 0.5), and it is not considered in detail in this paper as it is associated with the condensed phase instability.

One can estimate the change in the free energy from the particle center to its surface. This allows the physical interpretation of the introduced parameters of the model and the comparison of them with traditional energy characteristics which are used to describe the nucleation process.

Let us initially consider the layer--by--layer change in this free energy of a cylindrical drop. Let us return to the formula for the energy which we used to derive the equation of motion. It is as follows: $E(\mathbf{r}) = T + U$. Taking into account solution (9) we find that the kinetic energy is equal to the potential energy: T = U. This important result for the closed dynamic system is associated with the virial theorem for the finite motion, and in our case it is the test if this approach to the problem solution is correct. For the total full energy we have:

$$E(\rho) = T + U = 4A \left(\frac{a}{\rho}\right)^2 \frac{\rho^{2a}}{(1 + \rho^{2a})^2}.$$
 (26)

It follows from (26) that at a = 1 and r < 1 the equality E(r) = A is satisfied. In the case when B > 0 the cylinder surface energy tends to $Aa^2 \sim B$, and this limit is sharper the higher is the quantity B. Thus, just this parameter B can be associate to the parameter of the specific thermodynamic surface energy which occurs in the thermodynamic theories (of Gibbs, Tolman, etc.) on the understanding that the dimension of these energies is different.



Figure 6a: Dependence of energy on parameters *a* and ρ (see [9]).



Figure 6b: Dependence of energy on parameters *a* and ρ (see [8]).

A sharp rise in free energy (see Figures 6 a,b) depending on the parameter a is identified with the phase transition which takes place in the system in the case of infinitely small anisotropy (see for detail in [8, 9]). In order to determine the total energy of the particle assigned to the cylinder length unit an integral of $E(\mathbf{r})$ over the cylinder volume must be taken. Let us begin with a qualitative analysis of the model. Please note that for a particular case of a = 1 and B = 0 this integral must be equal to A (with an accuracy to the multiplier). Then there is no other energy in the system; here A is the only internal model energy of the system. In another limiting case some high value of a is sufficient for the total energy to tend to the anisotropy energy B. In the general case the total full specific energy (for the cylinder length unit) will be as follows:

$$W = 2\pi \int_{0}^{1} E(\rho)\rho d\rho = 8\pi a^{2} A \int_{0}^{1} \frac{\rho^{2a-1} d\rho}{(1+\rho^{2a})^{2}} = 2\pi a A.$$
(27)

In the Cahn--Hillard theory [9, 11] the activation barrier energy is in proportion to the geometric mean of two energy parameters:

$$E_c \sim \sqrt{AB}_{.(27a)}$$

Unlike the proposed theory the Cahn--Hillard theory is not scale--invariant, and the quantity B has a dimension of J/m^3 . In our case the integral formula derived from (11) for the activation energy has the same form, i. e., one may talk of the coincidence of these theories when calculating the mean activation energy (in the volume unit). Thus, we can deduce that the proposed theory coincide qualitatevely with the Cahn - Hillard theory. In frame Cahn - Hillard theory we obtain same analytic structures too [11]:



Figure 7: Schematics representations of the solution (25): (A), and (28): B) in the form of a domain wall of energy vectors.

The nanocylinder 2.

The previously introduced parameter a^2 (see (23)) can formally be less than a = 1 (in **Figure 5a**, this case is presented for a = 0.5). This case can correspond to negative anisotropy energy, which can, for example, prevent the creation of a nanoparticle. Technologically, it is possible to initiate the creation of a nanoparticle, but limit the growth a nanoparticle. Therefore, it is of interest to consider the case when:

$$B = -A, \qquad (29)$$

Such a physical situation can take place in the case when a nanoparticle nucleus with a size $\rho_0 < 1$ (for definiteness $\rho_0 = 0, 1$) has already been created, but its development to an equilibrium state with $\rho = 1$ is hindered by the created (artificially) anisotropy:

$$\frac{-A\sin^2\theta_a}{\rho^2}$$
. (30)

We believe that this anisotropy manifests itself only starting from some sizes corresponding to the value

$$\rho_0 = 0.1$$
 (31)

Then, for this case, the equation is linearized and greatly simplified:

$$\theta'' + \rho^{-1}\theta' = 0$$
. (32)

A particular solution for equation (32) can have the form (in our case, we use condition (32) to compare it with the solution in the form of formula (25)):

$$\pi - \theta = c \ln(\rho/\rho_0), (33)$$

where

 $c = \pi / (2\ln(1/\rho_0))$, (for $\rho > \rho_0$.).

Function graph $c \ln(\rho/\rho_0)$ shown in **Figure 8**.



Figure 8: Functions graphs of the solution (33).

Analytical remark

We can obtain from formulas (10), (12), (13) the asymptotic dependence

$$\tan\left(\varphi\right)\sim\frac{1}{\rho_r}, \quad (34)$$

where $\rho_r \sim \frac{r_z}{r_x}$ (see Figure 2). Radius r_z is analog r_1 and r_x is analog r_2 On the other hand, we got the formula (see (21)):

$$\tan\left(\frac{\theta}{2}\right) = \frac{1}{\rho}.$$
 (35)

We can obtain in classical theory (see Figure 1)

 $\tan\left(\frac{\theta}{2}\right) = \frac{H}{R}.$ (36)

These formulas have the same analytic structures. In frame Cahn--Hillard theory we obtain same analytic structures (form (28)) too [11]:

Conclusions

- a) With the decrease of the condensed phase in size the proportion of surface atoms increases thus increasing the influence of the interphase boundaries. At the same time the size dependence of surface tension is determined by the Tolman length i.e. the actual thickness of the interfacial (transition) layer.
- b) In the present work a formula for the surface tension dependence on the principal radii of curvature on an arbitrary surface (equation (6)) is correctly deduced. It is shown that curvature dependence of surface tension leads to a noticeable change in the equilibrium profile of sessile droplets on a flat non-wetting substrate. Note that the consequence resulted from the dependence of the surface tension on the surface curvature can be attributed to the capillary effect of the second kind **[8-10]**.
- c) At the end of the work there have been obtained the results associated with the van der Waals gradient theory which can be resumed in the following way. If in the formation of a nanoparticle there is only one energy form which plays the role of the exchange interaction *A* then the additive separation of the system energy into the surface energy and the nanoparticle volume energy in the context of the proposed model is incorrect. However, in this case one can introduce the average energy of the whole nanoparticle and, from simple geometric considerations, derive the Rusanov linear formula for the surface energy. Commonly, the Rusanov formula is assumed universally applicable. This fact is not confirmed when our model of the anisotropy energy is complicated.
- d) The concept of the anisotropy energy, introduced into the theory in the form of the proposed model as the Rapini modified potential, leads to the appearance of the surface energy. Please note that in the conventional Rapini potential there is no multiplier of the form $1/r^2$ [30]. The anisotropy energy can have the meaning of the double electric layer energy (in electrochemistry), besides, at the formation of very small equilibrium particles with the differentiated surface energy it should be increased the electric capacity of the nanosystem where this nanoparticle is formed. So, one can suppose that the nano--nucleation process can be efficiently controlled.
- e) The main findings that unite all the parts of the work are the dependences of the surface energy on the thermodynamic parameters of the system which make it possible to expand in some ways the vision of the capillary phenomena in nanosystems.

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