

## A TWO EQUATION MODEL OF BIOLOGICALLY REACTIVE SOLUTE TRANSPORT IN POROUS MEDIA

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**Summary:** Transport of biologically reactive dissolved solutes in a porous medium including a biofilm phase is a complex process involving a wide variety of scales (from the bacteria scale to the aquifer heterogeneity scale) and processes (hydrodynamic, physicochemical and biochemical). The objectives of this work are 1) to derive a two-equation macroscopic model for bio-reactive transport at the Darcy-scale from the pore-scale description using the volume averaging method and 2) to compare the results of this model with those of a recently developed one-equation model but limited to local mass equilibrium conditions.

### 1 INTRODUCTION

Mathematical modelling of transport in porous media of organic chemical species in presence of a bacterial population growing in the form of biofilms is an important area of research for environmental and industrial applications such as the treatment and the remediation of groundwater contaminated by organic pollutants, the study of bioreactor systems, ... Biofilms, which are composed of bacteria and extracellular organic substances, grow on the pore-walls of a porous medium. Biodegradable organic solutes are converted into biomass or other organic compounds by the bacterial metabolism. Accurate numerical models are needed in order to predict effectiveness of bioremediation methods or to optimize bio-processes occurring in engineering systems. This problematic has led to a broad literature (*e.g.*, [1]). One of the challenges in the development of such models is the variety of scales involved in the studied phenomena as shown in Figure 1. In this work, we focus on the

upscaling of the reactive transport processes from the pore scale to the Darcy scale.

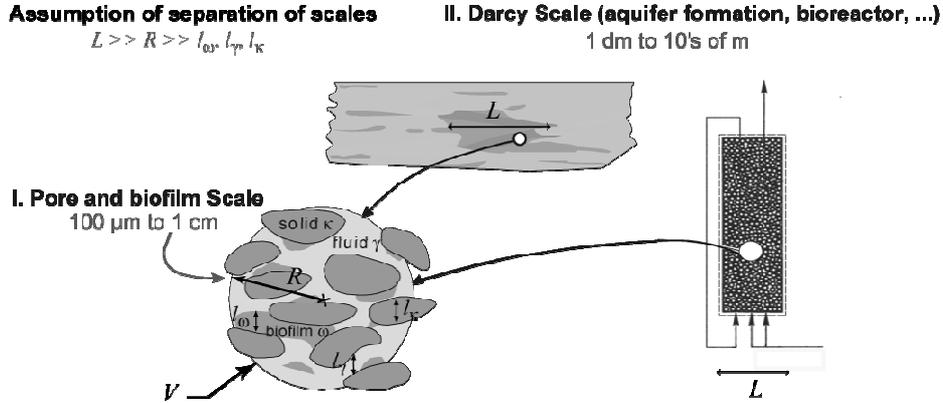


Figure 1. Pore scale and Darcy scale in a porous media with a biofilm phase.

We consider an organic solute, denoted  $A$ , which is classically transported by convection and diffusion in the fluid phase (where the reduced concentration field is denoted  $c_{A\gamma}$ ) and by diffusion within the biofilm phase (in which the reduced concentration field is denoted  $c_{A\omega}$ ). The reaction of biodegradation takes place in this last phase. A classical Monod kinetics expression will be adopted to describe the biological reaction (*e.g.*, [2] or [3]). Generally, biodegradation kinetics involves two chemical species: the source of carbon – organic solute  $A$  – and the electron acceptor – *e.g.*, dioxygen or nitrate, denoted  $B$ . For the sake of simplicity, we assume in this study that the electron acceptor is in large excess so that the consumption of solute  $A$  is governed by a simple Monod reaction term, even if such a simplification is not necessary regarding our theoretical development. The fluid and biofilm phases are assumed to be continuous and homogeneous except on the phase boundaries. The third phase, the solid phase, is considered to be passive relatively to the transport phenomena (no surface reaction). There is a feedback between solute transport and bacterial growth phenomena. In order to uncouple their governing equations, a quasi-steady assumption is considered based on the large difference between characteristic time scales of these processes (biomass growth is usually very slow compared to transport phenomena, *e.g.*: [4]). Under these assumptions, the pore-scale transport problem for solute  $A$  can be written as follows (all equations are presented in dimensionless form):

$$\frac{\partial c_{A\omega}}{\partial t} = \nabla \cdot (\mathcal{D}_A \nabla c_{A\omega}) - Da \frac{c_{A\omega}}{1 + c_{A\omega}/K_{A\omega}} \quad \text{in the } \omega\text{-phase}, \quad (1)$$

$$\frac{\partial c_{A\gamma}}{\partial t} + Pe \nabla \cdot (\mathbf{v}'_{\gamma} c_{A\gamma}) = \nabla^2 c_{A\gamma} \quad \text{in the } \gamma\text{-phase}, \quad (2)$$

with the boundary conditions:

$$\begin{aligned}
 -\mathbf{n}_{\omega\kappa} \cdot \mathcal{D}_A \nabla c_{A\omega} &= 0 \text{ at } A_{\omega\kappa}, & -\mathbf{n}_{\gamma\kappa} \cdot \nabla c_{A\gamma} &= 0 \text{ at } A_{\gamma\kappa}, \\
 -\mathbf{n}_{\gamma\omega} \cdot \mathcal{D}_A \nabla c_{A\omega} &= -\mathbf{n}_{\gamma\omega} \cdot \nabla c_{A\gamma} \text{ at } A_{\gamma\omega}, & c_{A\gamma} &= c_{A\omega} \text{ at } A_{\gamma\omega}.
 \end{aligned} \tag{3}$$

$A_{ij}$  represents the interface between the  $i$ -phase and the  $j$ -phase (each phase is identified by a greek letter - see Figure 1:  $\gamma$  for the fluid phase,  $\omega$  for the biofilm phase and  $\kappa$  for the solid phase).  $K'_{A\omega}$  is the dimensionless half saturation constant (reduced by the reference concentration  $c_0$ , the same used to obtain the reduced concentration fields  $c_{A\omega}$  and  $c_{A\gamma}$ ),  $\mathcal{D}_A$  is the ratio between the molecular diffusion coefficients within the biofilm phase and the fluid phase, and  $\mathbf{v}'_\gamma$  is the dimensionless velocity field (reduced by  $\|\mathbf{v}_\gamma\|$ ).

Here  $Pe$  and  $Da$  are the Péclet number and the Damköhler number associated to the transport phenomenon. They take the classical forms:

$$Pe = \frac{\|\mathbf{v}_\gamma\| l}{\mathcal{D}_{A\gamma}} ; \quad Da = \frac{\mu_{A\omega} \rho_\omega l^2}{\mathcal{D}_{A\gamma} K_{A\omega}} \tag{4}$$

in which  $\|\mathbf{v}_\gamma\|$  is the Euclidian norm of the velocity field  $\mathbf{v}_\gamma$ ,  $\mathcal{D}_{A\gamma}$  is the molecular diffusion coefficient in the fluid phase,  $\mu_{A\omega}$  is the maximum rate of substrate uptake,  $\rho_\omega$  is the microbial concentration (mass of active bacteria per unit of volume of biofilm),  $K_{A\omega}$  is the half-saturation constant and  $l$  is a reference length (e.g.: mean grains diameter, mean biofilm thickness, ...).

Upscaling of transport equations from the pore scale to the Darcy scale is conducted using the volume averaging method (e.g.: [5]). Microscale equations are averaged over a representative elementary volume  $V$ , which must satisfy the constraint of separation of scales (Figure 1). The resulting averaged quantities are the intrinsic averaged concentrations. In the  $\gamma$ -phase, it is defined as

$$\langle c_{A\gamma} \rangle^\gamma = \frac{1}{V_\gamma} \int_{V_\gamma} c_{A\gamma} dV \tag{5}$$

where  $V_\gamma$  represents the volume of the  $\gamma$ -phase contained in the averaging volume  $V$ . A similar definition holds for the  $\omega$ -phase. The resulting macroscopic conservation equations make appear some effective coefficients that remain dependent of the microscopic properties of the medium. In fact, some porescale 'closure problems' are mathematically stated relating the pore-scale physical characteristics to these effective properties. Heretofore, different one-equation averaged transport models have been rigorously derived but under restrictive assumptions: either for local mass equilibrium conditions [7] or for limiting transport mechanisms in non equilibrium conditions [8,9]. Actually, in the general case, the volume averaging process of the microscale transport equations leads to a two-equation model at the macroscale, where a separate upscaled equation is developed for each phase (e.g.: [10], [11]). Such a formulation is the subject of the current work. This description is the more complete

that can be obtained with such an approach, but a relatively high number of macroscopic effective coefficients has to be determined. In the following section we will briefly discuss the theoretical development of this *Two-Equation Model*, hereafter referred to as *TEM model*. In a second part, comparisons will be carried out between this upscaled model and one of the one-equation models derived previously [6]: the *Local Equilibrium Assumption (LEA) model* [7].

## 2 THEORETICAL DEVELOPEMENT

In the general case, concentration gradients may occur at the pore-scale in both phases, fluid and biofilm. In these conditions, the formalism of the volume averaging leads to a two-equation transport model at the Darcy scale. In addition to the classic constraint of separation of scales and in order to linearize the averaged reaction term, we need to assume that the spatial variations of deviation concentrations at the microscale (denoted  $\tilde{c}_{A\gamma}$  and  $\tilde{c}_{A\omega}$ ) are small compared to the value of the intrinsic averaged concentration ( $\langle c_{A\gamma} \rangle^\gamma$  and  $\langle c_{A\omega} \rangle^\omega$ ). Except of the reactive term, the problem stated with these assumptions is similar in essence with those studied by Cherblanc et al. [10] in the case of miscible transport of pollutant in heterogeneous porous media or by Ahmadi et al. [11] in the case of solute transport with adsorption.

The upscaling of the pore-scale problem (equations (1) to (4)) from the spatial operator defined in Eq. (5) and the use of averaging theorems (e.g. [5]) lead to unclosed average equations of transport defined at the Darcy scale, which still involve microscopic quantities (e.g. the microscopic reduced concentration fields  $c_{A\gamma}$  and  $c_{A\omega}$ ).

In order to close these transport equations, we need to establish a link between the microscale quantities and the macroscale quantities. The classical form of the closure assumptions associated with the volume averaging process is in such a case (e.g.: [9], [10]):

$$\tilde{c}_{A\gamma} = c_{A\gamma} - \langle c_{A\gamma} \rangle^\gamma = \mathbf{b}_{A\gamma\gamma} \cdot \nabla \langle c_{A\gamma} \rangle^\gamma + \mathbf{b}_{A\gamma\omega} \cdot \nabla \langle c_{A\omega} \rangle^\omega + r_{A\gamma} (\langle c_{A\omega} \rangle^\omega - \langle c_{A\gamma} \rangle^\gamma) \quad (6)$$

$$\tilde{c}_{A\omega} = c_{A\omega} - \langle c_{A\omega} \rangle^\omega = \mathbf{b}_{A\omega\gamma} \cdot \nabla \langle c_{A\gamma} \rangle^\gamma + \mathbf{b}_{A\omega\omega} \cdot \nabla \langle c_{A\omega} \rangle^\omega + r_{A\omega} (\langle c_{A\omega} \rangle^\omega - \langle c_{A\gamma} \rangle^\gamma) \quad (7)$$

where  $\tilde{c}_{A\gamma}$  and  $\tilde{c}_{A\omega}$  represent the concentration deviations and  $\mathbf{b}_{A\gamma\gamma}$ ,  $\mathbf{b}_{A\gamma\omega}$ ,  $r_{A\gamma}$ ,  $\mathbf{b}_{A\omega\gamma}$ ,  $\mathbf{b}_{A\omega\omega}$ ,  $r_{A\omega}$  are the closure fields, computed from solving three closure problems on a unit cell representative of the microscopic features of the medium.

Upon substituting these closure relations into the unclosed macroscopic equations allows one to define the following set of closed Darcy-scale transport equations:

*$\gamma$ -phase:*

$$\begin{aligned} \varepsilon_\gamma \frac{\partial \langle c_{A\gamma} \rangle^\gamma}{\partial t} + Pe \varepsilon_\gamma \langle \mathbf{v}_\gamma \rangle^\gamma \cdot \nabla \left( \langle c_{A\gamma} \rangle^\gamma \right) &= \nabla \cdot \left( \mathbf{D}_{A\gamma\gamma}^* \cdot \nabla \langle c_{A\gamma} \rangle^\gamma \right) + \nabla \cdot \left( \mathbf{D}_{A\gamma\omega}^* \cdot \nabla \langle c_{A\omega} \rangle^\omega \right) \\ + \nabla \cdot \left( \mathbf{d}_{A\gamma}^* \left( \langle c_{A\omega} \rangle^\omega - \langle c_{A\gamma} \rangle^\gamma \right) \right) - \alpha_A^* \left( \langle c_{A\omega} \rangle^\omega - \langle c_{A\gamma} \rangle^\gamma \right) &+ \mathbf{u}_{A\gamma\gamma}^* \cdot \nabla \langle c_{A\gamma} \rangle^\gamma + \mathbf{u}_{A\gamma\omega}^* \cdot \nabla \langle c_{A\omega} \rangle^\omega \end{aligned} \quad (8)$$

$\omega$ -phase:

$$\begin{aligned} \varepsilon_\omega \frac{\partial \langle c_{A\omega} \rangle^\omega}{\partial t} + \varepsilon_\omega Da \frac{\langle c_{A\omega} \rangle^\omega}{\langle c_{A\omega} \rangle^\omega + K_{A\omega}} &= \nabla \cdot \left( \mathbf{D}_{A\gamma\omega}^* \cdot \nabla \langle c_{A\gamma} \rangle^\gamma \right) + \nabla \cdot \left( \mathbf{D}_{A\omega\omega}^* \cdot \nabla \langle c_{A\omega} \rangle^\omega \right) \\ + \nabla \cdot \left( \mathbf{d}_{A\omega}^* \left( \langle c_{A\omega} \rangle^\omega - \langle c_{A\gamma} \rangle^\gamma \right) \right) + \alpha_A^* \left( \langle c_{A\omega} \rangle^\omega - \langle c_{A\gamma} \rangle^\gamma \right) &+ \mathbf{u}_{A\omega\gamma}^* \cdot \nabla \langle c_{A\gamma} \rangle^\gamma + \mathbf{u}_{A\omega\omega}^* \cdot \nabla \langle c_{A\omega} \rangle^\omega \end{aligned} \quad (9)$$

in which  $\mathbf{D}_{A\gamma\gamma}^*$ ,  $\mathbf{D}_{A\gamma\omega}^*$ ,  $\mathbf{D}_{A\omega\gamma}^*$  and  $\mathbf{D}_{A\omega\omega}^*$  are the effective dispersion tensors associated to the averaged concentrations  $\langle c_{A\gamma} \rangle^\gamma$  and  $\langle c_{A\omega} \rangle^\omega$  in the  $\gamma$ - and  $\omega$ -phases,  $\alpha_A^*$  is the mass exchange coefficient between phases and  $\mathbf{d}_{A\gamma}^*$ ,  $\mathbf{u}_{A\gamma}^*$ ,  $\mathbf{d}_{A\omega}^*$  and  $\mathbf{u}_{A\omega}^*$  are non-classical convective terms. All these effective parameters are functions of the above-mentioned closure variables. Indeed, three closure problems govern these variables and hence, the effective parameters which are defined as functions of integral values of these closure fields. For instance, we have:

$$\mathbf{D}_{A\gamma\gamma}^* = \frac{1}{V} \int_{A_{\omega\gamma}} \mathbf{n}_{\gamma\omega} \mathbf{b}'_{A\gamma\gamma} dA + \frac{1}{V} \int_{A_{\gamma\kappa}} \mathbf{n}_{\gamma\kappa} \mathbf{b}'_{A\gamma\gamma} dA - \frac{V_\gamma}{V} \langle \tilde{\mathbf{v}}'_\gamma \mathbf{b}'_{A\gamma\gamma} \rangle^\gamma + \frac{V_\gamma}{V} \mathbf{I} \quad (10)$$

Such problems are defined on the basis of the equations which govern the deviation fields  $\tilde{c}_{A\gamma}$  and  $\tilde{c}_{A\omega}$ . Briefly, they are obtained by subtracting the unclosed averaged equations to the microscopic equations, equations (1) to (4), and by introducing the closure relations, equations (6) and (7). As an example, one of the three closure problems is given below:

*Closure problem associated to the source term  $\nabla \langle c_{A\gamma} \rangle^\gamma$  :*

$\gamma$ -phase:

$$Pe \left( \tilde{\mathbf{v}}'_\gamma + \mathbf{v}'_\gamma \cdot \nabla \mathbf{b}'_{A\gamma\gamma} \right) = \nabla^2 \mathbf{b}'_{A\gamma\gamma} - \frac{1}{V_\gamma} \int_{A_{\omega\gamma}} \mathbf{n}_{\gamma\omega} \cdot \nabla \mathbf{b}'_{A\gamma\gamma} dA - \frac{1}{V_\gamma} \int_{A_{\gamma\kappa}} \mathbf{n}_{\gamma\kappa} \cdot \nabla \mathbf{b}'_{A\gamma\gamma} dA \quad (11)$$

$\omega$ -phase:

$$\nabla^2 \mathbf{b}'_{A\omega\gamma} - Da \frac{\mathbf{b}'_{A\omega\gamma}}{\left( K'_{eff,A}{}^{-1} + 1 \right)^2} = - \frac{1}{V_\omega} \int_{A_{\omega\gamma}} \mathbf{n}_{\gamma\omega} \cdot \nabla \mathbf{b}'_{A\omega\gamma} dA \quad (12)$$

$$\mathbf{n}_{\omega\kappa} \cdot \nabla \mathbf{b}'_{A\omega\gamma} = 0, \quad \text{at } A_{\omega\kappa}, \quad (13)$$

$$-\mathbf{n}_{\gamma\kappa} = \mathbf{n}_{\gamma\kappa} \cdot \nabla \mathbf{b}'_{A\gamma\gamma}, \quad \text{at } A_{\gamma\kappa} \quad (14)$$

$$\mathbf{n}_{\gamma\omega} + \mathbf{n}_{\gamma\omega} \cdot \nabla \mathbf{b}'_{A\gamma\gamma} = \mathbf{n}_{\gamma\omega} \cdot \mathcal{D}_A \nabla \mathbf{b}'_{A\omega\gamma}, \quad \text{at } A_{\gamma\omega} \quad (15)$$

$$\mathbf{b}'_{A\gamma\gamma} = \mathbf{b}'_{A\omega\gamma}, \quad \text{at } A_{\gamma\omega} \quad (16)$$

$$\langle \mathbf{b}'_{A\gamma\gamma} \rangle^\gamma = 0 \quad (17)$$

$$\langle \mathbf{b}'_{A\omega\gamma} \rangle^\omega = 0 \quad (18)$$

with:

$$\mathbf{b}'_{A\gamma\gamma} = \frac{\mathbf{b}_{A\gamma\gamma}}{l} \quad ; \quad \mathbf{b}'_{A\omega\gamma} = \frac{\mathbf{b}_{A\omega\gamma}}{l} \quad (19)$$

$$K'_{eff,A} = \frac{K'_{A\omega}}{\langle c_{A\omega} \rangle^\omega} \quad ; \quad \tilde{\mathbf{v}}'_\gamma = \mathbf{v}'_\gamma - \langle \mathbf{v}'_\gamma \rangle^\gamma \quad (20)$$

Notice that, in order to ensure unicity of the closure fields (here  $\mathbf{b}'_{A\gamma\gamma}$  and  $\mathbf{b}'_{A\omega\gamma}$ ), periodicity constraints are imposed at the boundaries. Finally, we have also supposed that  $\nabla \varepsilon_\gamma$  and  $\nabla \varepsilon_\omega$  are small in order to simplify the expression of the equations associated to the TEM model. Finally, the volume averaging method leads to the general form of the transport equations at the Darcy scale, and relates *explicitly* the values of the effective parameters to the pore-scale quantities through the so-called closure problems. Here the developed model is a two-equation model, which is the most general model which can be developed through this approach. In the next section, we will compare the results of this two-equation model with those obtained with a Darcy scale transport model also derived from the volume averaging technique but under more restrictive conditions: the Local Equilibrium Assumption model.

### 3 COMPARISON OF THE TWO-EQUATION MODEL WITH THE LOCAL EQUILIBRIUM ASSUMPTION MODEL

At this point, we have a general macroscopic model describing solute transport in a fluid-biofilm system which should *theoretically* include the validity domains associated to the LEA model. In order to justify this assumption and validate our development, we compare the effective parameters with those of this one-equation model, as developed in [7].

The local mass equilibrium assumption underlying the LEA model implies conditions close to thermodynamical equilibrium, i.e.:

$$\{c_A\} \approx \langle c_{A\gamma} \rangle^\gamma \approx \langle c_{A\omega} \rangle^\omega \quad (21)$$

Introducing this assumption and summing up macroscopic equations of the TEM model, Eqs. (8) and (9), we obtain:

$$(\varepsilon_\gamma + \varepsilon_\omega) \frac{\partial \{c_A\}}{\partial t} + \nabla (\varepsilon_\gamma \langle \mathbf{v}_\gamma \rangle^\gamma \{c_A\}) = \nabla (\mathbf{D}_{A,eff}^{*TEM} \cdot \nabla \{c_A\}) - \varepsilon_\omega \mu_{A\omega} \rho_\omega \frac{\{c_A\}}{\{c_A\} + K_{A\omega}} \quad (22)$$

where the effective dispersion tensor  $\mathbf{D}_{A,eff}^{*TEM}$  is given by:

$$\mathbf{D}_{A,eff}^{*TEM} = \mathbf{D}_{A\omega\gamma}^* + \mathbf{D}_{A\omega\omega}^* + \mathbf{D}_{A\gamma\gamma}^* + \mathbf{D}_{A\gamma\omega}^* \quad (23)$$

This macroscopic transport equation can be directly compared to the LEA model reminded below:

$$(\varepsilon_\gamma + \varepsilon_\omega) \frac{\partial \{c_A\}}{\partial t} + \nabla (\varepsilon_\gamma \langle \mathbf{v}_\gamma \rangle^\gamma \{c_A\}) = \nabla (\mathbf{D}_{A,eff}^* \cdot \nabla \{c_A\}) - \varepsilon_\omega \mu_{A\omega} \rho_\omega \frac{\{c_A\}}{\{c_A\} + K_{A\omega}} \quad (24)$$

Analogy between both models holds if  $\mathbf{D}_{A,eff}^{*TEM} = \mathbf{D}_{A,eff}^{*}$ . We compare (Figure 2) the evolution of the longitudinal component of these two effective dispersion tensors as a function of the Péclet number for the same porous medium than previously. Here we consider biochemical conditions consistent with the existence of a local mass equilibrium ( $Da = 0.1$ ,  $K_{A\omega} / \langle c_{A\omega} \rangle^\omega = 1000$ , and  $\mathcal{D}_A = 0.5$ ).

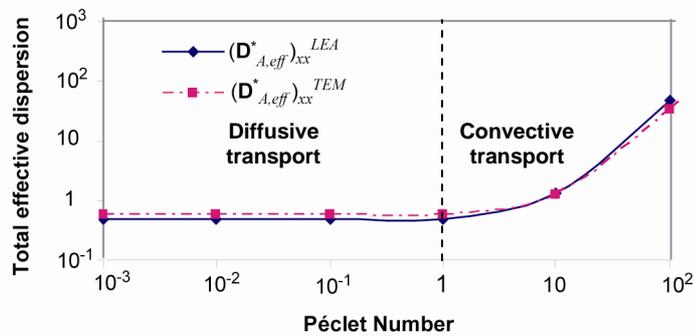


Figure 2. Comparison of the longitudinal dispersion obtained by the TEM model and the LEA model

The TEM model predicts a longitudinal dispersion which is slightly higher than the one given by the LEA model in the diffusive transport regime, while it gives a slightly lower dispersion in the convective regime. This slight discrepancy is due to the small - but non-zero - impact of the reactive term on the dispersion tensor in the TEM model (in the LEA model, this term is directly discarded in the writing of the closure problem associated to the dispersion tensor). However, the agreement between the two models is well satisfactory.

#### 4 CONCLUSION

In this paper, we have presented a two-equation macroscopic model describing solute transport in porous media hosting biofilms even for systems that are not at equilibrium. This study extends previous works conducted on this issue. A preliminary theoretical validation is presented based on a comparison between the TEM model and a one-equation model previously developed but under restrictive assumptions (local mass equilibrium). For this limit case, the one-equation behavior is well recovered by the TEM model. Note that an extensive numerical validation of the TEM model has been carried out by comparison between direct numerical simulations and upscaled simulations (data not shown) which confirm these observations and precise its validity domain.

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