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Estimation of drag force acting on spheres by slow flow and its application to a microfluidic device

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Abstract−The present study deals with the problem of determining drag force acting on spherical particles by slow flow through the particles in random arrays. Effective-medium theories using simplified models such as EM-I, EM-II, EM-III, and EM-IV are presented to predict the drag on the spheres in random arrays. These predictions are compared with numerical simulations. The EM-IV model in which the volume exclusion effect near the representative sphere is taken into account in defining the effective-medium is found to compare very well with the numerical simulations up to the volume fraction of spheres ϕ=0.5. In addition, Carman's correlation is given for comparison. This empirical correlation is shown to be in good agreement with the simulation results beyond ϕ=0.4. Therefore, it is found that selective use of EM-IV and Carman's correlation depending on ϕ is practically the best way to obtain accurate predictions of the drag for full range of ϕ. Finally, the estimations are compared with the previous experimental results for the gas pressure drop across a micropacked bed reactor. The comparison shows a reasonable agreement between the experimental results and the estimations by Carman's correlation.

Key words: Effective-medium Theory, Stokes Flow, Random Arrays, Microreactor, Micropacked Bed

INTRODUCTION

Stokes flow past randomly placed particles has been extensively studied through the rigorous calculation of the hydrodynamic interactions among the particles to finally yield the macroscopic transport properties such as drag coefficient, and hence permeability, for a particle-fluid system [Ganatos et al., 1978; Mazur and Saarloos, 1982; Bossis and Brady, 1984; Durlofsky et al., 1987; Brady and Bossis, 1988; Ladd, 1988; Sangani and Yao, 1988; Ladd, 1990; Mo and Sangani, 1994]. The rigorous calculations are complicated and the computation load becomes enormous when the number of particles is increased to 10^3 or 10^4. Therefore, it is necessary to develop relatively simple theories to predict the transport properties of a particle-fluid system. For this reason effective-medium theories have been developed. The effective-medium theories estimate one-particle conditionally averaged fields and hence the effective properties of the particle-fluid system by using a relatively simple model that captures some of the important multi-particle effects. The present study is concerned with providing the effective-medium theories for predicting the drag coefficient for Stokes flow through random arrays of particles. Four effective-medium models are presented and their estimations are compared with the numerical simulations. In addition, comparison is also made with Carman's correlation [1937] based on a capillary model which considers the space among closely packed particles as straight capillary tubes.

One of the recent applications of this Stokes flow analysis can be found in microfluidic devices since the fluid flows in the devices are usually within a low Reynolds number flow regime. The microfluidic devices for carrying out chemical reactions are called microreactors. In recent years the utilization of microreactors in practice has been rapidly increasing due to the advantage of high rate of heat and mass transfer [Jensen, 1999; Ehrfeld et al., 2000; Losey et al., 2001; Ajimera et al., 2001; Kusakabe et al., 2001; Choi et al., 2001; Wang et al., 2004; Sotowa et al., 2005]. Specifically, the present study deals with the case of a micro-packed bed reactor for phosgene synthesis to compare the experimental results by Ajimera et al. [2001] with the estimations for the gas pressure drop across the micropacked bed.

THEORY

We consider Stokes flow through a fixed bed of spherical particles randomly placed. The fluid velocity satisfies the equations of motion and the continuity equation given by

$$-\nabla p+\mu\nabla^2\mathbf{u}=0, \tag{1}$$

$$\nabla\cdot\mathbf{u}=0, \tag{2}$$

where p is the pressure in the fluid, **u** the fluid velocity, μ the viscosity of fluid. To model a fixed bed of spheres the present study assumes that the bed consists of a periodic array with each unit cell of the array containing N spheres whose positions are generated by using a specified spatial distribution law. The above boundary conditions must be satisfied on the surface of each sphere. The fluid velocity **u** must be spatially periodic and no-slip boundary condition is applied on the surface of each sphere. An additional constraint to be satisfied is

$$\frac{1}{\tau}\int_{V_f}\mathbf{u}\,dV=\mathbf{U}, \tag{3}$$

where **U** is the superficial fluid velocity through the bed, τ is the volume of the unit cell, and V_f is the volume occupied by the fluid within the basic unit cell.

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The above equations together with the boundary conditions can be solved by using a numerical method which computes the hydrodynamic interactions among spheres. Specifically, a multipole expansion method outlined in Mo and Sangani [1994] is used for the computation. Briefly, the method consists of writing a formal solution of Stokes equations of motion in terms of derivatives of a periodic fundamental singular solution of Stokes equations. This formal solution containing a number of undetermined coefficients satisfies the periodicity and the governing Stokes equations of motion. The coefficients are subsequently determined from expanding the formal solution around the surface of each sphere and satisfying the boundary conditions on the surface of spheres. This method finally yields the average force acting on a sphere, which is expressed by

$$F = 6\pi\mu a U K, \tag{4}$$

where K represents the non-dimensional drag on the sphere in a packed bed as a function of ϕ, the volume fraction occupied by the spheres of radius a. The pressure gradient by the flow through the bed is related to the force by

$$-\frac{dp}{dx_1} = nF = \frac{3\phi F}{4\pi a^3}. \tag{5}$$

where n is the number of spheres per unit volume of the bed. And the overall permeability k relating the mean flow to the mean pressure gradient is defined by

$$\mu\langle \mathbf{u}\rangle = -k\nabla\langle p\rangle. \tag{6}$$

Using Eq. (4) and Eq. (5), the permeability k is simply written as

$$\frac{k}{a^2} = \frac{2}{9\phi K}. \tag{7}$$

The present study is concerned with estimating non-dimensional drag K in a relatively simple manner using effective-medium theories in which the particle-fluid system is described as an effective-medium surrounding a representative particle. The effective-medium theories estimate one-particle conditionally averaged fields, and hence the properties of effective-medium by solving suitably averaged equations for a relatively simple model that captures some of the important multi-particle effects. Ensemble-averaging these equations subject to the presence of a sphere at **x** with its center at origin, **0**, yields

$$-\nabla p(\mathbf{x}|\mathbf{0}) + \mu(\mathbf{x})\nabla^2 \mathbf{u}(\mathbf{x}|\mathbf{0}) = 0 \tag{8}$$

with

$$\mu(\mathbf{x}) = \mu_f + (\mu^* - \mu_f)(1/\phi)\langle\chi\rangle_1(\mathbf{x}|\mathbf{0}). \tag{9}$$

Here, $\mu(\mathbf{x})$ is the viscosity at **x**, μ_f and μ^* are the viscosity of fluid and effective-medium, respectively, and χ is a phase indicator function whose value is unity when **x** lies inside a sphere and zero otherwise. $\langle\chi\rangle_1$ $(\mathbf{x}|\mathbf{0})$ is conditional average of χ that can be written as

$$\langle\chi\rangle(\mathbf{x}|\mathbf{0}) = \int_{|x-x'|\le a} P(\mathbf{x}'|\mathbf{0})\, dV_{x'} \tag{10}$$

where $P(\mathbf{x}'|\mathbf{0})$ is probability density for finding a sphere with its center at $\mathbf{x}'$ given the presence of a sphere at the origin. Note that χ approaches ϕ as $|\mathbf{x}|\to\infty$. The conditionally averaged pressure and velocity are required to approach, respectively, the unconditional averages as $|\mathbf{x}|\to\infty$.

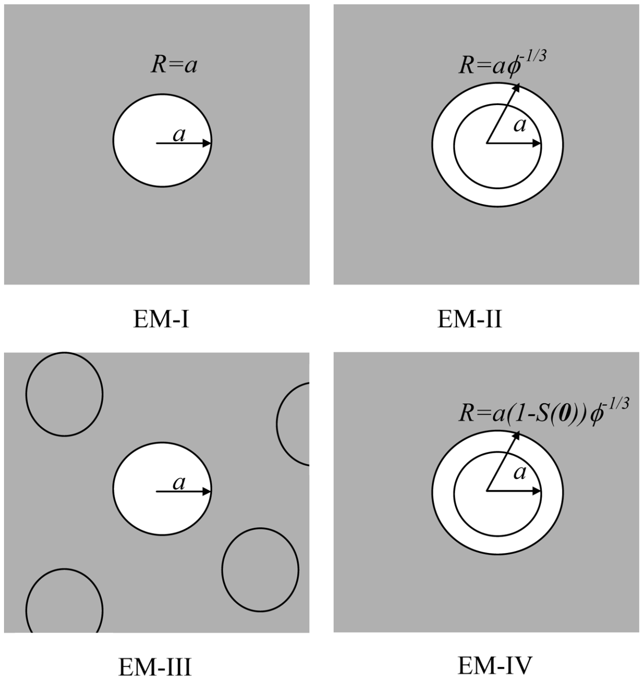

Fig. 1. Schematic diagrams for effective medium models.

In order to solve the conditionally averaged equations above, effective-medium models are presented and schematic diagrams for these models are shown in Fig. 1. The simplest is the model in which the effective-medium is taken to be outside of the representative particle at origin [Hill, 1965]. Thus the region for r>a, r being the distance from the origin, is considered as the effective-medium. This model is referred to as EM-I in present study.

More popular is the model in which the effective-medium begins at the larger distance $R=a\phi^{-1/3}$ [Hashin, 1983]. In this model, referred to as EM-II, a representative particle is surrounded by clear fluid up to the distance $R=a\phi^{-1/3}$ beyond which the particle-fluid system is considered as an effective-medium. Hence, for example, viscosity μ is taken to equal the suspending fluid viscosity μ_f in the region a<r<R and equal to effective viscosity μ^* of the effective-medium for r>R. Similarly, $\langle\chi\rangle_1$ is taken equal to zero in the region a<r<R and equal to ϕ in the effective-medium.

Later, Acrivos and Chang [1986] proposed an effective-medium model in which a representative particle is surrounded by an effective-medium whose properties such as density and viscosity are allowed to vary continuously, which is referred to as EM-III. The conditional average of the indicator function is determined by solving the Percus-Yevick equation [1958] for the radial distribution function for hard spheres. For suspensions in which the pair probability density is independent of the orientation of the pair, the volume integral in Eq. (10) can be reduced by using geometrical considerations shown in Fig. 2 to integration over s;

$$\langle\chi\rangle_1(\mathbf{x}|\mathbf{0}) = n\pi\int_{r-a}^{r+a} g(s)(2s - s^2/r - r + a^2/r)\, s\, ds, \tag{11}$$

where g is radial distribution function for hard spheres.

Since the determination of $\langle\chi\rangle_1$ and solution of EM-III are somewhat cumbersome, we need to develop a simple and accurate effec-

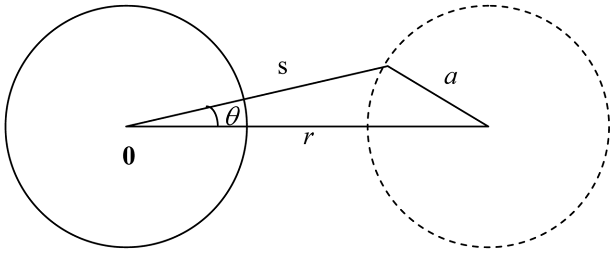

Fig. 2. Graphical representation of Eq. (11).

tive-medium model. This model, to be referred to as EM-IV, is similar to EM-II except the modification in defining the exclusion radius R given by

$$R = a\left(\frac{1-S(\mathbf{0})}{\phi}\right)^{1/3}. \tag{12}$$

Here $S(\mathbf{0})$ is the structure factor at zero wavenumber limit. The rationale for the choice of R is that there is a net depletion in the number density of spherical particles in the immediate vicinity of a particle since the particles are non-overlapping. The conditionally-averaged velocity satisfies the equations of motion for a single-phase flow in this exclusion region. Outside this region the particle-fluid medium is replaced by a medium whose properties are consistent with the average properties and average equations of motion for flow through a fixed bed of spherical particles, referred to as Brinkman medium in this case. The radius R is chosen so that the exclusion volume around a fixed particle equals that in the actual bed. Since the depletion of number density near the particle equals $P(\mathbf{r})-P(\mathbf{r}|\mathbf{0})$, we require that

$$\frac{4\pi R^3}{3} = \frac{1}{n}\int_{r>0}[P(\mathbf{r}) - P(\mathbf{r}|\mathbf{0})]dV_r. \tag{13}$$

The quantity on the right-hand side can be expressed in terms of zero wavenumber limit of the structure factor $S(\mathbf{0})$ to yield

$$\phi R^3 = a^3(1-S(\mathbf{0})), \tag{14}$$

where use has been made of the relation $\phi=4n\pi a^3/3$. The above choice for the exclusion radius R was made by Dodd et al. [1995] for solving the problem of determining the mobility of integral membrane proteins in bi-lipid membranes, modeled as suspensions of disks. Recently Koo and Sangani [2003] also used this exclusion radius R to solve the problem of determining the Sherwood number for longitudinal flow past arrays of cylinders. The zero wavenumber limit $S(\mathbf{0})$ can be evaluated by using the well-known Carnahan-Starling approximation:

$$S(\mathbf{0}) = \frac{(1-\phi)^4}{1+4\phi+4\phi^2-4\phi^3+\phi^4}. \tag{15}$$

Note that the exclusion radius R equals 2 in the limiting case of very small ϕ. R decreases as ϕ increases but remains greater than the radius of the particles. Once the exclusion radius R is determined, the conditionally averaged equations are simplified and readily solved. The velocity field and hence the drag on the test sphere at origin are determined by using a no-slip boundary condition on the test sphere and continuities of velocity and stress at the exclusion radius R.

RESULTS AND DISCUSSIONS

Table 1 shows the estimations for the drag acting on spherical particles by a single-phase flow using the effective-medium models. The permeability of the flow is also given as a function of ϕ in Fig. 3. The exact calculation results for the drag and the permeability by Koo [2005] are also presented for comparison. These results are found to be in excellent agreement with the previous results by Mo and Sangani [1994] and Ladd [1990]. These results for random arrays of spherical particles were obtained by averaging over twenty configurations generated by using a hard-sphere molecular dynamics code that employed 16 particles per unit cell. First, we see that EM-I model underpredicts the drag and shows a negative value at ϕ=0.6. The EM-II model gives estimations closer to the exact calculation results than EM-I does. However, its accuracy is much reduced near ϕ=0.6. This inaccuracy of the estimations by EM-I and EM-II at high volume fractions where the particles are nearly in contact is not observed in EM-III. But the estimations by EM-III are slightly lower than the exact calculations. EM-IV that is modified from EM-II to take the volume exclusion effect due to the presence of a representative particle at origin into account gives estimations in better agreement with the exact calculations for the drag acting spheres. It is also shown that EM-IV provides more accurate predictions for the drag than EM-II for $\phi\le 0.1$. Among the

Table 1. Non-dimensional drag K as a function of ϕ

ϕ	Exact calculations [Koo, 2005]	EM-I	EM-II	EM-III	EM-IV	Carman's correlation
0.01	1.28	1.26	1.48	1.40	1.28	0.10
0.1	2.86	2.40	3.12	2.77	2.76	1.37
0.2	5.59	4.21	5.61	5.27	5.30	3.91
0.25	7.1*	5.63	7.52	7.17	7.26	5.93
0.3	10.03	7.67	10.18	9.69	10.02	8.75
0.35	14.2*	10.78	14.06	13.11	14.02	12.74
0.4	19.58	15.86	19.91	17.87	20.04	18.52
0.45	28.1*	24.95	29.10	24.73	29.49	27.05
0.5	41.76	43.61	44.20	35.19	44.95	40.00
0.55		91.64	70.25	51.76	71.50	60.36
0.6	91.37	negative	220.9	79.25	223.9	93.75

*Data from Sangani and Mo [1994].

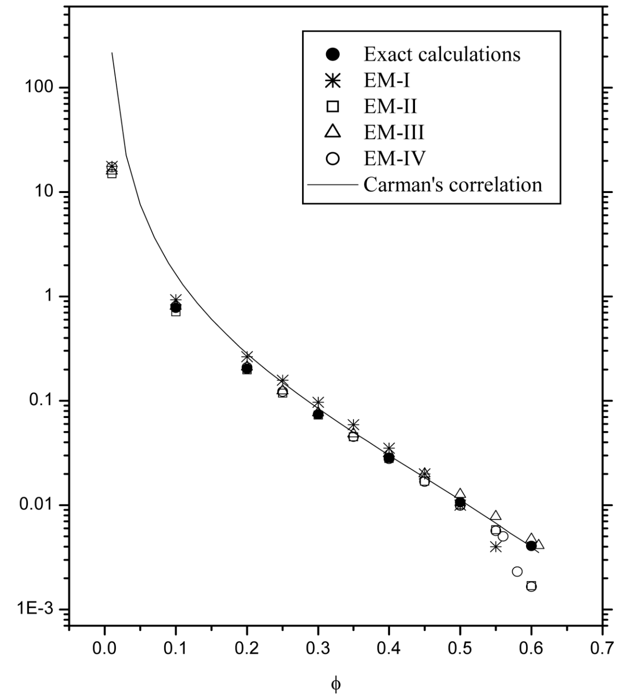

Fig. 3. Non-dimensional permeability k/a^2 as a function of ϕ.

above effective-medium models EM-IV is found to give the most accurate predictions for the drag acting on spheres. But the accuracy of the estimations by EM-IV is reduced for ϕ>0.5. Hence an alternative to the above effective-medium theories for predicting the drag at high ϕ may be to use a proper empirical correlation. Table 1 also shows the estimations by Carman's empirical correlation [1937], which is expressed by

$$\frac{F}{6\pi\mu aU}=\frac{10\phi}{(1-\phi)^3}. \tag{16}$$

We see that agreement between the estimation by Carman's correlation and the exact calculations is satisfactory for ϕ>0.4. Therefore, it is found that the combination of EM-IV for ϕ<0.4 and Carman's correlation for ϕ≥0.4 gives accurate predictions for the drag acting on spheres by Stokes flow for the full range of ϕ.

The above results are compared with the experimental results obtained by Ajimera et al. [2001] for phosgene synthesis using a micro-packed bed reactor loaded with active carbon particles of 60 μm in diameter as shown in Fig. 4. The dimension of the packed bed is 625 μm in width, 300 μm in depth, and 20 mm in length. Nitrogen gas was used for measuring the pressure drop across the bed. The range of the nitrogen gas flow rates for the experiment is 0-20 cm^3/min. The actual flow rates for the mixture of carbon monoxide and chlorine for phosgene synthesis are 4.5 and 8.0 cm^3/min. At these flow rates, the Reynolds numbers based on the particle radius are approximately 0.8 and 1.4, respectively. According to their paper, it is mentioned that they could obtain good agreement between the experimental results and the predictions using the Ergun equation [1952] for the pressure drop across the bed when the volume fraction of particles is fitted to be approximately 0.6. The Ergun equation given below is often used to predict the pressure drop in traditional packed beds.

$$\frac{dP}{dL}=-\frac{G}{\rho D_p}\frac{\phi}{(1-\phi)^3}\left[\frac{150\phi\mu}{D_p}+1.75G\right]. \tag{17}$$

Here dP/dL is the pressure gradient across the bed with the length L, G is the mass flow rate of the fluid, and D_p is the diameter of the particles. Since the Ergun equation is an empirical extension of Carman's correlation for the application to wider range of Reynolds number flows, the equation is reduced to Carman's correlation when the Reynolds number is small, but the coefficient 150 in Eq. (17) was corrected from 180 in Carman's original work for fitting empirical data. Hence a comparison is made between the estimations by Carman's correlation and the experimental results together with the exact calculations for the pressure drop through the micropacked bed by taking ϕ=0.6. Since Ajimera et al. [2001] report that the experimental results for the pressure drop follow the Ergun equation

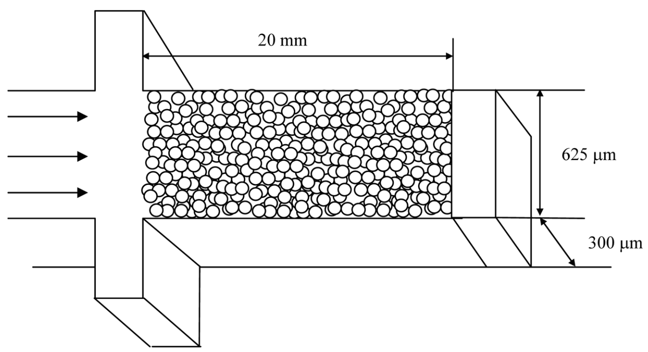

Fig. 4. Schematic diagram of a micropacked bed reactor [Ajimera et al., 2001].

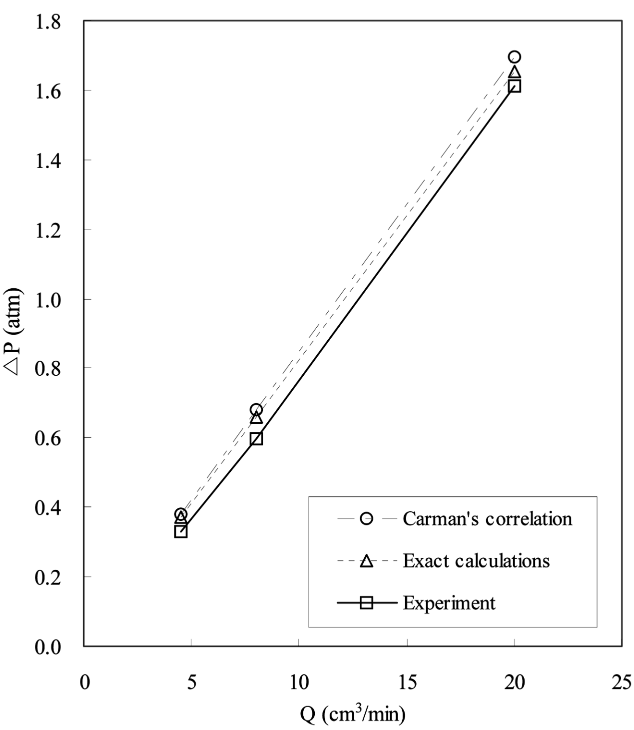

Fig. 5. Pressure drop across a micropacked bed as a function of a gas flow rate, Q.

with ϕ taken to be approximately 0.6, the pressure drop from using the Ergun equation is considered as the experimental results for the comparison. Fig. 5 shows that there is a good agreement among the experimental results, the predictions by the correlation and the exact calculations. However, the actual volume fraction of particles in the experiment is not known and small difference in ϕ can cause the calculations to digress from the experimental results. Thus it may not be precise to estimate the pressure drop across the bed by using the above calculations with a fitted value of ϕ. But, it is also known that ϕ=0.6 is a reasonable choice for the closely packed spheres in random arrangement from the literature. [Chong et al., 1971] Therefore, it is concluded that Carman's simple correlation is reasonably accurate for estimating the pressure drop by slow flow through the particles in a micropacked bed.

SUMMARY

The drag acting on spherical particles by slow flow past the particles in random arrays is determined by using effective-medium theories. Four effective-medium models, i.e. EM-I, EM-II, EM-III, and EM-IV, are presented for predicting the drag acting on spheres. Among these models EM-IV in which volume exclusion effect around a representative sphere is taken into account is found to be the most accurate, but the accuracy of the predictions using the effective-medium theories is reduced at high ϕ. Using Carman's empirical correlation based on a capillary model can be an alternative to the effective-medium theories at high ϕ. It is shown that the estimations of the drag by Carman's correlation compare very well with the exact calculations beyond ϕ=0.4. Hence accurate predictions for the drag acting on spheres by slow flow for wide range of ϕ can be obtained in a relatively simple manner by the selective use of EM-IV and Carman's correlation depending on ϕ. Comparison is also made with the experimental results for the pressure drop across a micropacked bed. It is shown that there is a reasonable agreement between the experimental results and Carman's correlation for the pressure drop across a micropacked bed.

NOMENCLATURE

a : radius of particle
D_p : diameter of particle
g : radial distribution function
G : mass flow rate of fluid
k : overall permeability
n : number density of particles
p : pressure in the fluid
$P(\mathbf{x}|\mathbf{0})$: probability of finding a particle at $\mathbf{x}$ given a particle at origin $\mathbf{0}$
r : radial distance from the center of the particle at origin
$S(\mathbf{0})$: structure factor at zero wavenumber limit
$\mathbf{u}$: velocity of the fluid
$\mathbf{U}$: superficial velocity of the fluid through the bed
$\mathbf{x}$: position vector
χ : particle phase indicator function
$<\chi>_1$: pair probability density, conditional ensemble average of χ
ϕ : volume fraction of the particles
μ_f : viscosity of fluid
μ^* : effective viscosity
τ : unit cell volume

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