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ON NON-FICKIAN HYPERBOLIC DIFFUSION

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Abstract: Fick's law expresses the proportionality of solute flux with respect to concentration gradient. Similar relations are given by Darcy's law for the fluid flow in porous media, Ohm's law for the electric flux and Fourier's law for heat transfers. When introduced in the corresponding balance equations, these flux laws yield diffusion equations of parabolic character. Different attempts have been made to obtain hyperbolic equations so as to point out propagative phenomena. This was done by adding a time derivative flux term to the flow law. In this paper, we focus on solute transport. Two possible non-Fickian diffusion cases are addressed. We firstly investigate diffusion in fluids by a mechanistic approach. Secondly, we study the macroscopic diffusion law in composite materials with large contrast of diffusion coefficient. We show that the diffusion law obtained yields hyperbolicity for drastically short characteristic times or non-propagative waves.

1. INTRODUCTION

Isotropic Fick's law for diffusion of solute in fluids is given by

$$\vec{J} = -D\vec{\nabla}c \,, \tag{1}$$

where $\vec{J}$ is the solute flux, D is the molecular diffusion and c is the solute concentration. When introduced into the solute balance equation

$$\vec{\nabla}.\vec{J} = -\frac{\partial c}{\partial t} \tag{2}$$

equation (1) yields a diffusion equation of parabolic character

$$\vec{\nabla}.D\vec{\nabla}c = \frac{\partial c}{\partial t} \,. \tag{3}$$

In such parabolic second-order differential equations, information propagates at an infinite velocity, a property which is common to all diffusion processes, i.e., to Fick's, Fourier's, Ohm's or Darcy's processes. To overcome this "non-physical" behaviour, modifications are introduced into these laws so as to render equation (3) hyperbolic. First attempts concerned Fourier's law, see MAXWELL [11], CATTANEO [4], VERNOTTE [14], [15], CHESTER [5], VOLZ et al. [16], MOYNE and DEGIOVANNI [12] and DUHAMEL [7] for a more complete bibliography. Ohm's law is also addressed in a similar way, see, e.g., CUEVAS et al. [6]. Darcy's law poses less difficulty since it is a momentum balance equation whose extension to dynamics is easily demonstrated, LEVY [10], AURIAULT

[1]. That yields to Biot's law, BIOT [3], which shows added mass phenomenon. For solute transport, studies are not numerous. SCHEIDEGGER [13] proposed a hyperbolic diffusion equation, which corresponds to a modified Fick's law, HASSANIZADEH [8]. Let us consider the isotropic form of this hyperbolic diffusion law

$$\vec{J} + A\frac{\partial \vec{J}}{\partial t} = -D\vec{\nabla}c\ , \tag{4}$$

where time $A > 0$ characterizes together with D the solute transport. Relation (4) is similar to the Cattaneo–Vernotte law for heat flow, CATTANEO [4], VERNOTTE [15]. When introduced into the solute balance equation, such a non-Fickian law yields a hyperbolic equation which enables us to point out finite velocity propagation phenomena: the time A is responsible for inertial effects

$$\vec{\nabla}.D\vec{\nabla}c = \frac{\partial c}{\partial t} + A\frac{\partial^2 c}{\partial t^2}\ . \tag{5}$$

The form of transport equation (4) is often justified as being a consequence of material heterogeneity, DUHAMEL [7]. Anyhow, MOYNE and DEGIOVANNI [12] have pointed out that a transport equation of propagative type could not be recovered in one-dimensional composite materials. In this paper, we present two attempts to recover such a non-Fickian law (4). In part 2, we investigate transient solute diffusion in liquids by a mechanistic approach following Einstein's diffusion theory. We demonstrate that such a relation (4) is valid, but for very short characteristic transport times. Finally, in part 3, we discuss the possible non-Fickian character of solute flow through a composite material with high contrast of diffusion coefficients. For this purpose, we refer to the paper by AURIAULT [2], where the macroscopic equivalent heat flow law is obtained by upscaling the physics at the heterogeneity scale.

2. NON-FICKIAN HYPERBOLIC DIFFUSION IN LIQUIDS

The molecular diffusion is related to the mobility b by the Einstein relation

$$D = kTb\ , \tag{6}$$

where k is the Botzmann constant, $k \approx 1.38 \cdot 10^{-23}$ J K^{-1} and T is the absolute temperature.

2.1. QUASI-STATIC DIFFUSION COEFFICIENT

Consider a solute particle moving at a constant velocity v in a stagnant fluid. The fluid exercices on the particle a force F which is related to v by

$$F = \frac{v}{b}. \tag{7}$$

In the case of spherical particle, b is given by the Stokes formula

$$b = \frac{1}{6\pi\mu a}, \tag{8}$$

where μ is the viscosity of the fluid and a is the particle radius. Therefore, we obtain the quasi-static diffusion coefficient in the form

$$D_0 = \frac{kT}{6\pi\mu a}. \tag{9}$$

2.2. TRANSIENT DIFFUSION

When the particle velocity is not a constant and depends on the time t, relation (7) takes the form (see LANDAU and LIFCHITZ [9], §24)

$$F = 2\pi\rho a^3\left(\frac{1}{3}\frac{\mathrm{d}v}{\mathrm{d}t} + \frac{3\mu}{\rho a^2}v + \frac{3}{a}\sqrt{\frac{\mu}{\pi\rho}}\int_{-\infty}^{t}\frac{\mathrm{d}v}{\mathrm{d}\tau}\frac{\mathrm{d}\tau}{\sqrt{\mathrm{t}-\tau}}\right). \tag{10}$$

Using (7) to obtain the value of b in the transient case and (6) for D yields the following non-Fickian law

$$-D_0\vec{\nabla}c = \frac{\rho a^2}{3\mu}\left(\frac{1}{3}\frac{\mathrm{d}\vec{J}}{\mathrm{d}t} + \frac{3\mu}{\rho a^2}\vec{J} + \frac{3}{a}\sqrt{\frac{\mu}{\pi\rho}}\int_{-\infty}^{t}\frac{\mathrm{d}\vec{J}}{\mathrm{d}\tau}\frac{\mathrm{d}\tau}{\sqrt{\mathrm{t}-\tau}}\right). \tag{11}$$

Relation (11) represents the correct generalization of quasi-static Fick's law (1) to dynamics. It is different from (4) due to the presence of memory effects that are represented by the last term in the right-hand member. The memory function assumes the form

$$M(t) = \sqrt{\frac{\rho a^2}{\pi\mu t}}, \tag{12}$$

and its characteristic time is given by

$$T_m = \frac{\rho a^2}{\pi\mu}. \tag{13}$$

2.3. ESTIMATION OF THE DOMAIN OF VALIDITY OF NON-FICKIAN RELATION (11)

Assume that the fluid is water, $\mu = 10^{-3}$ kg m^{-1} s^{-1}, $\rho = 10^3$ kg m^{-3}. For $a = 10$ Å we obtain $T_m \approx 3 \times 10^{-13}$ s. For $a = 10^{-6}$ m we have $T_m \approx 3 \times 10^{-7}$ s.

On the other hand, the right-hand member of relation (11) introduces two-dimensionless numbers P and Q. Noting T_c the characteristic time of the process yields

$$\mathrm{P} = \frac{\left|\frac{1}{3}\frac{\mathrm{d}\vec{J}}{\mathrm{d}t}\right|}{\left|\frac{3}{a}\sqrt{\frac{\mu}{\pi\rho}}\int_{-\infty}^{t}\frac{\mathrm{d}\vec{J}}{\mathrm{d}\tau}\frac{\mathrm{d}\tau}{\sqrt{\mathrm{t}-\tau}}\right|} = \frac{\pi}{9}\sqrt{\frac{T_m}{T_c}},$$

$$\mathrm{Q} = \frac{\left|\frac{1}{3}\frac{\mathrm{d}\vec{J}}{\mathrm{d}t}\right|}{\left|\frac{3\mu}{\rho a^2}\vec{J}\right|} = \frac{\pi}{9}\frac{T_m}{T_c}.$$

We can then point out three different behaviours:

- $T_c >> T_m$. Relation (11) reduces to Fick's law (1).
- $T_c = O(T_m)$. The flow law is given by (11) with its three terms in the right-hand member. This is a non-Fickian law, but different from (4).
- $T_c << T_m$. The two last terms in (11) are negligible. This is a non-Fickian law which yields a hyperbolic behaviour as in (4), but without the first term in the right-hand member.

However, as can be seen above, T_m is very small. Therefore, the two last behaviours are likely not to exist. Remark that it is relation (10) (where dynamics is considered) that gives the two last behaviours the hyperbolic character.

3. MEMORY EFFECTS IN COMPOSITE MATERIALS

Consider a periodic composite material of the period Ω, made of two constituents of diffusion coefficients D_1 and D_2 which occupy the volumes Ω_1 and Ω_2, respectively. The interface is Γ. Medium 1 is connected, whereas medium 2 is connected or not (see the figure, where medium 2 is not connected). Both constituents follow Fick's law (1) and equation (3). The characteristic size of the heterogeneities or of the period is l, whereas the characteristic size of the macroscopic sample or of the macro-

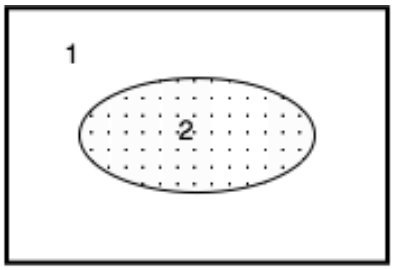
scopic solute flux is L and we assume a separation of scales: $l << L$. When $D_1 = O(D_2)$, the macroscopic equivalent behaviour is Fickian (see AURIAULT [2], who investigated heat flow in composite materials): the macroscopic equivalent description is given by

$$\vec{\nabla}.D^{\text{eff}}\vec{\nabla}c = \frac{\partial c}{\partial t},$$

where D^{eff} is the effective diffusion tensor which depends on both constituents 1 and 2 and on the geometry at the fine scale. $D_2 = O(D_1 l/L)$ or $D_2 = O(D_1(l/L)^3)$ are particular cases of the previous one.

We consider here the case where medium 2 is much less conductive than medium 1

$$\frac{D_2}{D_1} = O\left(\frac{l}{L}\right)^2 << 1.$$



Schematic view of a period (2D case)

The problem of a macroscopic equivalent behaviour was investigated by AURIAULT [2] for heat transfer by using the method of multiscale asymptotic expansions. The reader is referred to this paper for details of the upscaling process.

The upscaling process yields the following equivalent macroscopic behaviour, at the first order of approximation (the model is valid within a relative error $O(l/L)$)

$$\vec{\nabla}.D_1^{\text{eff}}\vec{\nabla}c = \frac{\partial c}{\partial t} - \int_{-\infty}^{t} K^{\text{eff}}(t-\tau)\frac{\partial^2 c(\tau)}{\partial \tau^2}\,\mathrm{d}\tau. \tag{14}$$

In relation (14), c represents the concentration in medium 1 and D_1^{eff} is the effective diffusion tensor of the composite material, when considering medium 2 as a non--diffusive. The macroscopic solute flux is shown to be

$$\vec{J} = -D_1^{\text{eff}}\vec{\nabla}c. \tag{15}$$

The memory function $K^{\text{eff}}(t)$ is obtained as the inverse Fourier transform of $\bar{k}/i\omega$, where

$$\bar{k}(\omega) = \frac{1}{|\Omega|} \int_{\Omega_2} k(\omega) \mathrm{d}V \tag{16}$$

and the complex periodic function $k(\omega)$ is the solution of the following boundary value problem in Ω_2

$$\vec{\nabla}.D_2 \vec{\nabla} k = i\omega(k-1), \tag{17}$$

$$k = 0 \quad \text{on } \Gamma.$$

Illustrations for $\bar{k}$ are given by AURIAULT [2] for layered media and for circular cylindrical or spherical inclusions. Important properties of the real and imaginary parts of $\bar{k}$ are that both are positive and allow us to verify (AURIAULT [2]) the following inequalities

$$\bar{k} = \bar{k}_1 + i\bar{k}_2,$$

$$0 < \bar{k}_1 \le n, \quad 0 < \bar{k}_2 \le n, \tag{18}$$

$$n = \frac{|\Omega_2|}{|\Omega|},$$

$$\lim_{\omega \to 0} \frac{\bar{k}_2}{\omega} > 0.$$

Relation (14) can be seen as describing a Fickian process in the presence of a source term represented by the second term in the right member: the macroscopic equivalent medium is described by solute concentration in medium 1 and Fick's law (15). Medium 2 plays the role of a solute reservoir connected to medium 1. Medium 1 is responsible for the macroscopic diffusion flux. However, the form of the source term in (14) encourages us to look for an equivalent description in the form of non-Fickian relation (5). This can be done by considering solute flow at the constant frequency ω. Relation (14) then becomes

$$\vec{\nabla}.D_1^{\mathrm{eff}} \vec{\nabla} c = (1-\bar{k}_1)\frac{\partial c}{\partial t} - \frac{\bar{k}_2}{\omega}\frac{\partial^2 c}{\partial \tau^2} \mathrm{d}\tau. \tag{19}$$

We recover (5), but with a different solute capacity $1-\bar{k}_1 < 1$ and with $A = -\bar{k}_2/\omega < 0$. Therefore, differential equation (19) is elliptic, of the nature different from (5) which is hyperbolic. There is no finite velocity propagation in the composite material. This result was also obtained by a different approach in MOYNE and DEGIOVANNI [12] for one-dimensional heat flow. On the contrary to section 2, no

dynamics is present at the heterogeneity scale nor is introduced later. This explains why we do not recover a propagative process at the macroscopic scale. It is likely that some dynamics should be introduced somewhere so as to obtain a propagation process. Remark that the upscaling method in use is rigorous in the sense that the macroscopic description is obtained from the fine scale description, without any prerequisite as it is usual with other methods.

4. CONCLUSION

Two attempts to justify the non-Fickian Cattaneo–Vernotte law for solute diffusion have been made in a view to enable finite velocity propagation of solute perturbations. By analyzing molecular diffusion through its mechanistic Einstein definition, it was shown that such a hyperbolic description is valid, but for transient solute flows with unrealistic very short characteristic times. In composite material with diffusion coefficients of similar or of very different order of magnitude, we did not recover the Cattaneo–Vernotte law, but either a parabolic or an elliptic phenomenon.

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