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Numerical investigation of generalized Graetz problem in circular tube with a mass transfer coupling between the solid and the liquid

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ABSTRACT

The unsteady diffusion equation in a tube and the advection/diffusion equation in laminar flow in a liquid of a solute are theoretically established. The most important parameters are the migration strength α and the so-called Graetz number Gr. The former dimensionless number is the product of the partition coefficient of the solute at the solid/liquid interface, the ratio of the tube thickness to the interior radius and the ratio of diffusion coefficients of the solute in the liquid and in the solid. The Graetz number Gr is the ratio of the diffusion time scale in the liquid based on the interior radius of the pipe to the advection time over the tube length. The problem is applied to the plasticizer migration from a polyvinyl chloride material to a liquid with the safety food applications in mind. The migration of the plasticizer is solved numerically for various tube sizes, flow conditions and partition coefficients of the plasticizer in the liquid. For comparison, the numerical results obtained in static condition are also provided.

The average plasticizer concentration in the liquid behaves following two main regimes as a function of the migration strength. When α is much smaller than one, the average plasticizer concentration in the liquid is an algebraic function of the axial coordinate, z, at the power two third and increases linearly with time. Conversely, when α is much larger than one, the average plasticizer concentration is linear as a function of axial distance and increases as a function of the square root of time. Moreover, the concentration is much smaller of few orders of magnitude in dynamic condition than in static working showing that in the context of food contamination the dynamic approach is more appropriate and relevant for demonstration of compliance with the safety authorities requirements.

1. Introduction

Heat transfer between a fluid flowing through a solid tube is commonly used to design heat mass exchanger devices, see the recent book of Zhang [1]. Since the first contributions of Graetz [2,3], the heat transfer between a solid and a liquid has been extensively studied and summarized in the book of Shah and London [4]. According to [5,6], the analogy between heat and mass transfer in the limit of mass diffusion of a solute in weak concentration in a solvent can be invoked meaning that the same physics can be applied to migration issues.

The last situation occurs in the food delivery through tubes made from thermoplastic matrices involved in milk producing and transforming, vending machines among others. The contact between food and its container leads to a release of additives in the food. According to safety requirements, articles or materials in contact with food have to be in compliance dictated by the food safety authorities. Two quantities have to be mainly determined to prove the compliance: the former is the overall migration corresponding to the total amount of material migrating in the food per unit of surface of the container in contact with food, measured in mg/dm^2. The latter is the specific migration corresponding to the migration of particular substances by mass unit of food in the container. The European Union (EU) regulation 10/2011 [7] sets the Overall Migration Limit (OML) equal to 10 mg/dm^2. For substances without Specific Migration Limit (SML), a generic SML equal to 60 mg/kg shall apply. However, this value can be reduced for certain substances like for polydimethylsiloxane, for which the SML is equal to 6 mg/kg, for instance.

The compliance of a plastic material should be measured using food simulants under specified test conditions. The specification of EU regulation has been based on static conditions, for which food

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simulant is in contact with the container during a certain time. Nevertheless, for deliveries through hoses, liquid food is in motion. Consequently, the mass transfer with solid/liquid interfaces exhibits different behaviors in comparison with the static situation. Additives are transported with the food limiting the accumulation. Mass transfer involves mass diffusion in the tube and advection of the liquid food. Consequently, the migration process in dynamic conditions has to be studied in more details, which is the aim of this work. The important questions which arise are the following: What are the main differences between migration without or with motion? What will be the consequences in terms of safety requirements? In order to reply to those questions, this article is devoted to the numerical modelling of the dynamic and static conjugated migration problems. It is noteworthy to say that the problem addressed here is general and can be also applied to the heating or the cooling of a tube.

The conjugated problem has been addressed for the first time for heat transfer applications by Mori et al. [8] and latter by Faghri and Sparrow [9] in the situation of high conduction in the solid meaning that only the steady state regime has been studied. A special attention to axial conduction has been investigated in [10–12] but once again in steady state regime both in the liquid and in the solid. More recently, Zhang et al. [13] achieved a numerical work of the conjugated problem by solving the Navier–Stokes equations and the two energy equations. A parametric study was carried out on the ratio of thermal diffusivity and on the ratio of the thickness of solid tube to the interior diameter of the pipe.

In the framework of safety food requirements, numerical simulations are generally used with a simple 1-D geometry. The migration model is generally based on the development provided by Crank [14] to determine the migration of a substance well stirred in the solution (food simulant). The process is only limited by diffusion in the PVC sheet since the solution is seen as a perfect diffuser. Begley et al. [15] summarized the main steps to build up and validate a migration estimation model. They compiled a large amount of data in order to determine the "worst-case" migration level. The method requires the knowledge of diffusion coefficients of additives in the packaging material and of the partition coefficients which define the chemical equilibrium at the food/container interface. Vitrac et al. [16] described the migration model based on the local diffusion process requiring the mass transfer coefficient in the food simulant depending on fluid motion of the simulant. They identified diffusion, partition, and mass transfer coefficients by an optimized calculation. Brandsch et al. [17] summarized how the mathematical modeling can be used to evaluate the migration limits.

The numerical simulation of migration process in a cylindrical tube has been seldom investigated. Let us quote nevertheless, works achieved in the framework of water distribution for which the diffusion of antioxidant from hoses to hot-water has been investigated by Smith et al. [18] and more recently by Dear and Mason [19]. The diffusion of chlorine from water to the tube is also taken into account and reacts with the antioxidant substance following an irreversible reaction. Nevertheless, the water motion is not addressed in previous works. Mittelman et al. [20] studied the same problem for which an exact solution was proposed when chemical reactions are neglected. Otherwise, a numerical simulation was established with a finite difference technique.

The dynamic condition of migration of plasticizer requires a special attention which is the main purpose of the present work. Indeed, apart from the flowing behavior, the migration process is studied for hoses for which the dilution can be strongly limited for small tube. Moreover, the diffusion process exhibits particular behaviors like for instance the tube curvature. In order to study the migration, a theoretical model is developed and solved numerically to describe the migration in dynamic conditions. Since the aim of this present study is to compare the migration in static and dynamic conditions, the conjugated problem of diffusion between two media at rest is also presented and solved numerically. A special attention will be done on the amount of additive migrating in the liquid at a function of time.

In the following, the problem statement of the dynamic migration is described in the Section 2 in which a dimensionless formulation is built pointing out the relevant dimensionless numbers. Section 3 is devoted to the presentation of results and discussion in which the effects of the geometry, flow rate and plasticizer solubility are addressed before to conclude in Section 4. Appendix A presents the conjugated static migration problem. Details of the numerical methods both for the dynamic and static problems are provided in supplementary material of this article.

2. Problem statement of the dynamic conjugated migration

Although the migration process is addressed both in static and dynamic conditions, only the conjugated problem of mass diffusion in dynamic condition is presented below. Since the static problem has been considered elsewhere, the presentation in this last situation is reported in Appendix A.

Only one substance is considered in the problem. In the solid tube, the mass concentration of the plasticizer is written C_p and C_f in the liquid. The diffusion coefficients of the plasticizer are denoted D_p and D_f in the solid and in the liquid respectively. The plasticizer concentration in the pipe is in large amount in order to increase the flexibility.

As it is well known in materials science, the diffusion of plasticizer in solid material depends upon the plasticizer concentration [21]. A decrease of plasticizer concentration changes the temperature of the glass transition of the PVC material, decreasing drastically the diffusion coefficient [22]. It is worthy to note that all over this work, the diffusion coefficients are assumed in first approximation constant over the time of migration process. This assumption means that the mass transfer will be limited to the first times in order to avoid a large decrease of plasticizer in the PVC material. In fact, the time scale of diffusion in the PVC material is inversely proportional to the diffusion coefficient. The typical value of the time scale of diffusion for a tube of one millimeter in thickness is larger than 2700 h by taking a diffusion coefficient D_p equal to 10^{-13} m^2/s which is a typical value for a plasticizer in flexible PVC tube [23]. The time is very high in comparison with the typical times of migration experiments which do not exceed few tens of minutes meaning that the decrease of plasticizer concentration stays low. Note also that by assuming a constant value for the diffusion coefficient, the migration process is treated in the worst situation, since when the diffusion coefficient decreases as a function of time, a skin layer acting as a functional barrier against migration is created. Moreover, the aim here is to compare with the static situation for which the same assumption is used. The change of diffusion coefficient as a function of the plasticizer concentration should not modify the main conclusions of the present investigation.

In the liquid, the diffusion coefficient is determined assuming that the additive concentration in the liquid is sufficiently small in order to apply the Wilke and Chang's correlation [24]. A numerical value will be provided at the beginning of Section 3.

Fig. 1 summarizes the problem statement where typical profiles of C_p and C_f are sketched. The tube length is L, and interior and exterior radii are r_i and r_e respectively. The liquid flows with a volumetric flow rate equal to Q and has a constant density, ρ and dynamic viscosity, μ. In the following, the flow regime is assumed laminar meaning that the typical Reynolds number is less than