Numerical model to solve impurities’ migration in water pipes

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Abstract. We present a novel numerical method based on finite differences to solve the problem of impurities’ migration from a hollow core multilayer cylinder filled with a known liquid medium. We expose the numerical method and the developed application. The validation is performed by calculating the migration in a configuration which allows an analytic solution. Applications are foreseen in modeling impurities’ migration from modern multilayer water pipes into drinking water.

1. Introduction
The use of polyethylene (PE) pipes in drinking water distribution networks and household installations causes a contamination of the consumers’ tap water which is a major concern of water pipes producers. Two types of PE pipes are used in the drinking water network: high density PE (HDPE) pipes in the distribution mains and service pipes, and/or cross linked PE (PEX) in the household installations as hot and cold water pipes. More than 100 different organic compounds are found to migrate into water from polyethylene materials like HDPE and PEX (see for example [1, 2]).

The model presented here is similar to that used to calculate migration of additives from food packaging polymers into the food [3] developed in one dimension and taking into account the European Commission’s recommendations [4]. In these cases the main chemical parameters of importance are the migrant’s diffusion coefficient in the polymer and its partition coefficient at the interface. The concentration of the migrant in this model is assumed to have no boundary layer resistance between packaging and food [5]. The basic difficulty, however, lies in the composition of the media from which the migration takes place, i.e. food packaging or water pipes, as these media are nowadays constituted of multilayer materials. This structure introduces new parameters for the modeling like the diffusion coefficients of the migrant in different layers and the partition coefficient between two adjacent layers as defined by the ratio between migrant solubility in the two layers.

One should stress that the case of diffusion in a multilayer system of cylindrical geometry is present not only in water pipes but also in food packaging. Indeed, in many cases, liquid food like beverages, oil, fruit juices, etc. are packed in PET bottles and the calculation of migrated substance from the bottles into the food can be influenced to a certain extent by this geometry.

The paper is organized as follows: in Section 2 we present the actual problem to be solved numerically, then in Section 3 we validate the used numerical procedure and present the obtained
results. Finally, we conclude and refer to further developments and applications of the model.

2. Exposition of the Problem

The equation of diffusion in cylindrical geometry has the form:

\[ \frac{\partial c}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( r D \frac{\partial c}{\partial r} \right). \tag{1} \]

A numerical solution of the diffusion equation is obtained by performing the following steps: (i) define a numerical grid; (ii) use a numerical method (for example Lagrange polynomials) to approximate finite differences at mesh points; (iii) discretize the diffusion equation using the method of finite differences; (iv) solve the resultant system of algebraic equations. For our case the algebraic equations have the form:

\[ c_{n+1}^i - c_n^i = D \delta t \cdot F \left[ \theta \left( A c_{n+1}^{i-1} - B c_{n+1}^i + E c_{n+1}^{i+1} \right) + (1 - \theta) \left( A c_{n}^{i-1} - B c_{n}^i + E c_n^{i+1} \right) \right], \tag{2} \]

where \( c_n^i \) denotes the migrant concentration at mesh point \( i \) and time \( n\delta t \), while \( A, B, E, F \) are constants resulting from the geometry of the system and the implementation of the Lagrange polynomials for the derivatives.

Discretization on a numerical grid of the diffusion equation for the case of multilayer systems is not trivial at the interfaces between adjacent layers. Due to the non-unitary value of \( K \) partition coefficient (in general), the concentration profile of the migrant is not continuous at the interface between layers. A suitable method to treat this numerical difficulty is to implement the fictitious point method. Both the general finite difference and fictitious point methods are presented in Ref. [3] for planar geometry.

The method presented above was further developed in a computer program application which was tested by modeling a case in which analytical solution can be derived. The results of the validation of the method will be presented in the following section.

3. Method Testing and Results

Our goal is to test and validate the numerical method sketched in the previous section. For this purpose we need to find a particular case which can be treated analytically. In his book Crank [6] discusses the problem of migration in a cylinder and gives the solution under different conditions. The closest case to our practical situation is the migration of impurities from a well-stirred solution surrounding concentrically a solid cylinder. In other words the solution is playing the role of the hollow core pipe while the inner cylinder plays the role of water. The initial concentration of migrant in the solution is \( C_0 \) and the water is initially free from migrant. This problem can be treated analytically and would correspond to the two-layer case in the numerical simulation. The amount \( M_t \) of migrant in the cylinder after time \( t \) is expressed as a fraction of the corresponding amount \( M_\infty \) after infinite time by the relation:

\[ \frac{M_t}{M_\infty} = 1 - \frac{\gamma_3}{\gamma_3 + \gamma_4} \exp \left[ 4 \gamma_3^2 \frac{Dt}{r^2 \alpha^2} \right] erf \left[ \frac{2 \gamma_3}{\alpha} \left( \frac{Dt}{r^2} \right)^{1/2} \right] - \frac{\gamma_4}{\gamma_3 + \gamma_4} \exp \left[ 4 \gamma_4^2 \frac{Dt}{r^2 \alpha^2} \right] erf \left[ -\frac{2 \gamma_4}{\alpha} \left( \frac{Dt}{r^2} \right)^{1/2} \right], \tag{3} \]

where \( \alpha = V_{pipe}/(K \cdot V_{water}) \) with \( K \) partition coefficient, \( \gamma_3 = \frac{1}{2} \left( 1 + \alpha \right)^{1/2} + 1 \) and \( \gamma_4 = \gamma_3 - 1 \). The inner and outer radii of the pipe are \( r \) and \( R \), respectively. As shown in Ref. [6] this solution is valid only for small values of \( Dt/r^2 \) and \( \alpha \) moderate. This limitation will become visible also on our results.
In Fig. 1, we represent the temporal variation of the total amount of migrant in the water for different values of the partition coefficient at the pipe/water boundary. A value $K$ here means that the concentration of the migrant in the water just at the interface with the pipe is $K$ times that in the pipe at the interface with the water. Other parameters of the model are: $r = 1 \text{ cm}$, $R = 1.01 \text{ cm}$, $C_0 = 1000 \text{ mg/kg}$, $D = 10^{-12} \text{ cm}^2/\text{s}$.

One can observe a reasonable agreement of the numerical results (dashed lines) with the curves corresponding to Eq. (3) for partition coefficient $K = 1$ and 10. However, we remark that this agreement is limited to times less then 10000 hours in the case of partition coefficient $K = 0.1$. We can read from the graph that for very large times ($t > 10^8$ hours) the analytical formula completely fails. Indeed, according to Crank [6] Eq. (3) works only for small $Dt/r^2$, which is $3.6 \cdot 10^{-4}$ for $t = 10^8$ hours, but for $t = 10^9$ hours is as high as 3.6. On the other hand, the analytical condition to keep $\alpha$ "moderate" is not so well-defined. In our case $\alpha = 0.00201$ for $K = 10$, $\alpha = 0.0201$ for $K = 1$ and $\alpha = 0.201$ for $K = 0.1$. Based on Fig. 1, this latter value proves to be no more "moderate".

Another very important constraint to obtain reasonable agreement between analytical results using Eq. (3) and the numerical results is to fulfill the condition of well-stirred solution of migrant in the pipe. We implement this constraint numerically by setting the diffusion coefficient of migrant inside the pipe to a value many orders of magnitude larger than its diffusion coefficient in water. Results presented on both figures are obtained with $D_{\text{water}} = 10^{-12} \text{cm}^2/\text{s}$ and $D_{\text{pipe}} = 10^{-7} \text{cm}^2/\text{s}$. Obviously, these values are completely unphysical. In reality diffusion coefficients governing migration of impurities in water are much larger then the corresponding values for diffusion in the PE pipes. The only reason we have yet chosen these unrealistic values was to fit in the constraints for the validity of the analytical formula, such that direct comparison can be performed and the numerical results can be tested.

On Fig. 2, we represent numerical results for the spatial profile of the migrant concentration in the vicinity of the layer boundary for different values of $K$ after different times of migration. One can remark the continuity of the concentration curve at the water/pipe interface when $K = 1$, as expected. In the case when $K = 0.1$ we have a finite jump in the migrant concentration at the interface. Regardless of the amount of time elapsed from the beginning of the migration the concentration-jump at the interface is conserved at the fixed value $c_{\text{water}}/c_{\text{pipe}} = K = 0.1$ validating the correctness of the numerical model.
The generalization of the numerical method to simulate migration in multilayer systems is straightforward, while we have seen the serious limitations of the analytical treatment.

4. Conclusions
In this paper we exposed the problem of migration of impurities from hollow pipes into their cylindrical inner core. The most important practical occurrence of this configuration is the case of PE water pipes and any kind of foodstuff in cylindrically shaped plastic packaging. Experimental measurements involving real systems are very expensive and time consuming. The availability of a suitable model, however, can overcome both of these difficulties. Here we presented such a model, tested and validated it by implementing a very simple case which can be treated analytically and we were able to directly compare the analytical and numerical results.

As a conclusion we can state that the presented numerical model is suitable to simulate migration of impurities through multilayer systems with cylindrical geometry. In comparison with the limited possibilities of analytical treatment this numerical method has several major advantages: (i) its validity range is not limited in time, nor in what concerns any other parameter of the real system (diffusion and partition coefficients, layer thickness, etc.) and (ii) it can be very easily generalized to arbitrary number of cylindrical layers which is the real situation in case of PE water pipes and polymer food packaging materials as well.

The next step of this research will be to simulate multilayer cases with realistic parameters, to fit them with experimental data, thus we hope to obtain good estimates of quantities which are very difficult to be measured experimentally.

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