Modelling the diffusion of lead into drinking water

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Abstract

The use of lead in pipe networks, and the subsequent lead emissions into drinking water, is now a major concern to the European Union Member States. Computer simulation offers the possibility for achieving a greater understanding of the issues that relate to plumbosolvency control. An analytical and a numerical scheme for the diffusion of lead into drinking water within a pipe during a stagnation period are described. These have been validated by experimental work. Three models have been investigated to simulate the flow of water through a pipe. These flow models have been used in conjunction with the equation of mass transfer to simulate the concentration of lead at the tap. This has been used within a Monte Carlo framework in order to simulate a zone of properties. The zonal output data obtained consists of failure rates at specified lead standards. This data can be utilised to investigate and prioritise plumbosolvency control options, and substantially overcomes the severe limitations of sampling. Preliminary results from the zonal model compare well with published DWI data.

1 Introduction

Lead is a toxic metal which, if absorbed, can lead to serious health problems. The use of lead in pipe networks, and the subsequent lead emissions into drinking water, is now a major concern to the European Union Member States. The recently revised European Union Drinking Water Directive [1] requires Member States to reduce the levels of lead in their drinking water. To achieve this, water can either be chemically treated or the lead pipes can be replaced or a combination of the two. The cost of replacing all the lead pipes across Europe
has been estimated at 70 Billion Euros [2]. Due to the logistics involved it is not feasible to evaluate the amount of lead in drinking water for every house in a water supply zone. Instead, a handful of random samples are taken; however, it is questionable how representative this is. Without adequate quantification of compliance to the new directive, prioritisation of corrective actions must become speculative and the ability to justify actions and demonstrate their success must be weakened [3]. Computer simulation offers the possibility for achieving a greater understanding of the issues that relate to plumbosolvency control. With this aim in mind, research has been focused on constructing a computational model that will assist water engineers to assess the problem and to make informed decisions.

2 Mass transfer

For simplicity it is assumed that the pipework at a single property consists of a length of lead pipe connected to a length of non-lead (copper) pipe which is then connected to a tap. Two models have been investigated to simulate the transfer of lead from the lead pipe into the water, namely the exponential model and the more complex diffusion model.

2.1 Exponential model

The exponential model assumes that the mass transfer of lead, from the internal surface of a pipe into the water within the pipe, is a function of the initial mass transfer rate $M$ (μg/m²/s), and the equilibrium concentration of lead in the water, $E$ (μg/l). The parameters $M$ and $E$ define the plumbosolvency (a measure of the extent of lead dissolution) of the water [3]. $M$ and $E$ can be calibrated by reference to "over-night standing" and "30-minute stagnation" samples from properties with known pipe-work characteristics and from plumbosolvency propensity tests in the laboratory [4]. Assuming the source of lead, $S$, is directly proportional to the degree of saturation of lead with respect to $E$, i.e. $S \propto (E - c)/E$, it can be shown [3] that the concentration, $c$, after a stagnation period of length $T$ can be written as:

$$c = E - (E - c_0)e^{-\frac{AMT}{VE}} \quad (1)$$

where $c_0$ is the initial concentration at the start of the no-flow period and $A$ (m²) and $V$ (m³) are the internal area and volume of the pipe, respectively.

2.2 Diffusion - analytical solution

The diffusion model is based upon the mass transfer equation, which describes how the concentration of lead $c$ varies through the pipe (co-ordinate system $x$, $r$, $\phi$) and over time with respect to the principal velocities of the fluid ($u$, $v$, $w$) and
the coefficient of diffusion $D \text{ (m}^2/\text{s)}$. Assuming no flow and no variations in the angular direction and along the pipe, the mass transfer equation can be written

$$\frac{\partial c}{\partial t} = D \left[ \frac{\partial^2 c}{\partial r^2} + \frac{1}{r} \frac{\partial c}{\partial r} \right]$$

Eqn (2) is solved subject to the boundary conditions

$$c = c_0 = E \text{ at } r = r_b \text{ for } t \geq 0$$
$$c = f(r) \text{ } 0 < r < r_b \text{ for } t = 0$$

The solution of eqn (2) subject to eqn (3) describes how the concentration varies across the pipe cross-section and is given by [5]

$$c = c_0 \left[ 1 - \frac{2}{r_0} \sum_{n=1}^{\infty} \frac{1}{\alpha_n J_1(r_0 \alpha_n)} \exp(-D \alpha_n^2 t) \right]$$
$$+ \frac{2}{r_0^2} \sum_{n=1}^{\infty} \exp(-D \alpha_n^2 t) \frac{J_0(r \alpha_n)}{J_1(r \alpha_n)} \int_0^r f(r') J_0(r' \alpha_n) dr'.$$

where $f(r)$ is the initial concentration distribution across the pipe. $J_0$ and $J_1$ are Bessel functions and $\alpha_n$ are the positive roots of

$$J_0(r_0 \alpha_n) = 0$$

This solution is more accurate and provides more detail than the exponential model, as the concentration is now a function of $r$.

2.3 Diffusion – numerical solution

To accurately model the diffusion of the existing lead in the water within the non-lead pipe during stagnation, it is necessary to develop a numerical solution since the Dirichlet boundary condition imposed by the analytical solution is not appropriate. In a numerical solution a more appropriate Neumann boundary condition can be applied in the case of the non-lead pipe.

The pipe is treated as a cylinder which is discretised along the axis $(i, i = 1, N)$, and in the radial direction $(j, j = 1, J)$. The concentration of lead in the pipe $c_j^n$ is defined as

$$c_j^n = c(j \Delta r, n \Delta t)$$
where $\Delta r$ is the increment in the radial direction ($\Delta r = r_{j+1} - r_{j}$) and $\Delta t$ is the increment in time. The diffusion equation, eqn (2), may then be rearranged and written in finite difference form as

$$C_j^n = C_j^{n-1} + \Delta tD \left[ \frac{C_{j+1}^{n-1} - 2C_j^{n-1} + C_{j-1}^{n-1}}{\Delta r^2} + \frac{C_{j+1}^{n-1} - C_{j-1}^{n-1}}{2r_j\Delta r} \right]$$

(7)

with boundary conditions

$$\frac{\partial C}{\partial r} = 0 \text{ at } r = r_0, \text{ for } t \geq 0$$

(8)

$$C = C_0 = E \text{ at } r = r_0, \text{ for } t \geq 0$$

(9)

$$\frac{\partial C}{\partial r} = 0 \text{ at } r = 0, \text{ for } t \geq 0$$

(10)

This solution, eqn (7), is valid for $0 < r_j < r_0$ and is 1st order accurate in time and 2nd order accurate in space. The Neumann boundary condition, eqn (8), is applied at the copper pipe while the Dirichlet boundary condition, eqn (9), is applied at the lead pipe. We cannot use a Neumann boundary condition for the lead pipe because the rate of dissolution is not constant. Note that it is also necessary to apply a symmetry condition, eqn (10), at the middle of the pipe [6].

### 2.4 Experimental work

Experimental data was obtained in order to substantiate the lead dissolution response predicted by the diffusion model. Data obtained will reveal the plumbosolvency characteristics of the water investigated as well as enabling the diffusion model to be more fully calibrated. Least-squares curve fitting of the experimental results with the diffusion model has been carried out in order to calibrate the diffusion model and to investigate the overall closeness of fit of the data (Figure 1). From this plot it is clear that the experimental results can be fit very closely to the predicted diffusion model values. A diffusion coefficient of $D = 3.55E-10 \text{ m}^2/\text{s}$ has been obtained from this calibration. This compares favourably when calibrating to the 30 minute stagnation concentration of the exponential model where the diffusion coefficient was found to be $D = 4.46E-10 \text{ m}^2/\text{s}$ [6].
Figure 1: Concentration variation against stagnation time.

3 Flow in pipe

3.1 Plug flow

Lead enters the water through the process of diffusion when the tap is closed as described above. Once the tap is open, the dissolved lead moves along the length of the pipework, being carried by the movement of the water. In this case, the governing equations for the fluid motion are the Navier-Stokes equations. Typically, the Reynolds number is the order of 8,000 for 'tap open' situations and hence the flow is assumed fully turbulent. However, for flexibility a choice of three different flow models can be used, namely plug-flow, laminar and turbulent.

In the Plug-flow model the fluid is assumed to be ideal and is treated as inviscid. As the name suggests, plug-flow implies that the fluid has constant velocity across the cross-section of the pipe and progresses down the pipe as a 'plug' of fluid. Thus in this case \( u = \text{constant} = U \), and \( v = 0 \). Though not as accurate as the other two models it is computationally very quick.

3.2 Laminar and turbulent flow

If we assume radial symmetry and \( v = 0 \) the Navier-Stokes equations can be reduced to

\[
\frac{\partial p}{\partial x} = \mu \left[ \frac{1}{r} \frac{\partial u}{\partial r} + \frac{\partial^2 u}{\partial r^2} \right]
\]  

(11)
which leads to the solution of laminar flow (where $Q$ is the volumetric flow rate):

$$u = \frac{2Q}{\pi r_0^4} \left( r_0^2 - r^2 \right) \quad (12)$$

For turbulent flow a simple power law is a good approximation. The most widely used power law is the One-Seventh-Power Law, this leads to:

$$u = \frac{Q}{0.817 \pi r_0^2} \left( \frac{r_0 - r}{r_0} \right)^{1/7} \quad (13)$$

### 3.3 General solution

If the exponential model is used to simulate lead transfer during flow, then the mass transfer equation can be taken to be

$$\frac{\partial c}{\partial t} + u \frac{\partial c}{\partial x} = S \quad (14)$$

To solve eqn (14) for any of the velocity profiles, a simple finite difference procedure is used. The pipe is treated as a cylinder which is discretised along the axis ($i, i = 1, N$), and in the radial direction ($j, j = 1, 4$). Eqn (14), which is equivalent to the conservation of mass of lead within an element $(i,j)$, leads to the equation

$$\frac{\partial c_{i,j}}{\partial t} = \frac{u_{i,j}}{\Delta x_i} \left( c_{i-1,j} - c_{i,j} \right) + A_i \sum_j M_{i,j} \left( \frac{E - c_{i,j}}{E} \right) \quad (15)$$

where $c_{i,j}, V_{i,j}$ and $A_i$ are the concentration of lead in element $(i,j)$, the volume of element $(i,j)$ and the internal surface area of element $i$ at the wall of the pipe, respectively and $Q$ and $t$ are the flow rate and time, respectively [6]. It is now a simple matter of substituting $u_{i,j}$ for a constant (plug-flow) or for eqns (12) or (13) (laminar and turbulent flow respectively).

Upon implementing eqn (15) we are able to compute concentration variation over space and time as illustrated (Figure 2).
4 Zonal model

4.1 Daily average simulation

For a particular property it is clear that these mass transfer and flow models can be utilised to compute the average concentration of lead from the tap over the course of one day assuming pipework configuration and water usage information is known. This is what is called the ‘single house model’. However, in the water industry the extent of the problem of lead in drinking water needs to be quantified for a whole water supply zone, typically comprising more that 10,000 individual properties.

A zonal simulation involves executing the single house model for each simulated property within the simulated zone. Firstly, the number of properties in the zone and the percentage of properties estimated to be supplied through lead pipe-work are specified. Attributes for each simulated property such as pipe lengths, volume of water used per day and pattern of water used are ascribed randomly from a given distribution function. This distribution function is assumed to be log-normal in the case of pipe lengths and normal in the case of volume of water used. The water usage patterns are pre-defined and ascribed equally.

The principal output from the zonal model is a summary of the percentage of houses in the zone that fail to meet the 10, 25 and 50 µg/l lead standards, on the basis of the daily average concentrations of the lead emissions across the zone.
4.2 Random daytime sampling

In real life it is clearly not feasible to measure the daily average concentration for every single house in a zone because of the logistics involved. Consequently there is no real world data of this type that can be used to validate the results from the model. However, a sampling model has been developed alongside the daily average concentration model, which simulates the process of Random Day Time (RDT) Sampling. This is the standard procedure carried out by water companies to obtain an idea of the zonal lead compliance. It involves taking a one-litre sample at a random time from a random property in the zone. Typically, one random daytime sample will be collected every week over the course of one year [6].

Results from the sampling model compare favourably to real world data from random daytime sampling [7] for a wide range of water types thus validating the model (Table 1).

### Table 1: Computed RDT results vs. actual RDT results.

<table>
<thead>
<tr>
<th>Plumbosolvency Propensity</th>
<th>Computed Average Failures (%)</th>
<th>RDT Failures (%)</th>
<th>Actual Reported (DWI) RDT Failures (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>&gt;10mg/l</td>
<td>&gt;25mg/l</td>
<td>&gt;50mg/l</td>
</tr>
<tr>
<td>High</td>
<td>29.50</td>
<td>16.90</td>
<td>7.60</td>
</tr>
<tr>
<td>Moderate</td>
<td>20.10</td>
<td>7.40</td>
<td>1.80</td>
</tr>
<tr>
<td>Low</td>
<td>1.70</td>
<td>0.00</td>
<td>0.00</td>
</tr>
</tbody>
</table>

4.3 Using the model

The sampling model can be used to predict the expected failure rates for existing zones. Also, by simulating a number of RDT surveys for the same zone, it is possible to predict the likely spread of results. This can be used to evaluate the reproducibility and suitability of the RDT sampling procedure itself.

Both daily average and sampling models have been used to investigate the effects of selective lead-pipe replacement as well as the effects of corrective water treatment. Corrective water treatment involves the dosing of phosphate before water distribution. This effectively lowers the plumbosolvency of the water by forming a scale on the inside of the pipe, acting as a physical barrier between the water and the lead-pipe. Water treatment is simulated simply by altering the input parameters for the zone. Typical RDT output data, which illustrates the effects of water treatment is shown (Figure 3). The X% reduction refers to the reduction in plumbosolvency (defined by $M$ and $E$). This plot also demonstrates the variability in failure rates occurring against the 10μg/l standard over the 100 surveys simulated.
Figure 3: Number of failures against the 10μg/l standard.

5 Conclusions

Two models have been investigated to simulate the transfer of lead into the water within a pipe during a stagnation period. An analytical solution as well as a numerical solution has been developed for the more complex diffusion model. These models have also been substantiated by experimental results, which have allowed the diffusion models to be calibrated more fully.

Three models have been investigated to simulate the flow of water through a pipe. These flow models have been used in conjunction with the equation of mass transfer to simulate the concentration of lead at the tap. This single house model has been used within a Monte Carlo framework in order to simulate a zone of properties. The random daytime sampling procedure can be simulated on this zone and the corresponding failure rates are output. The model is also capable of computing the daily average concentration for every single property in the zone and output the relevant statistics. Preliminary results compare well with published DWI data.

As well as overcoming the severe limitations of traditional RDT sampling, the model can be used to investigate selective lead-pipe replacement and the effects of water treatment.
References


