GAS DIFFUSION IN HEAT INSULATED PLASTIC PIPE WITH INNER AND OUTER BARRIER LAYERS

By Evgeniy Devyatkin, Igor Gvozdev, Sergey Shalyapin

P U foam with a closed porosity structure is extensively used for heat insulation of pre-insulated plastic pipes. PU foams have good heat insulating properties due to low heat conductivity of gases trapped in the cells. One of the disadvantages of PU is its λ-ageing (deterioration of heat insulating properties over time) due to gradual substitution of gases with low heat conductivity into air components with higher heat conductivity [1–4]. Oxygen penetration into PU foam initiates its thermal oxidative degradation which can compromise the integrity of heat supply pipelines. Polymer barrier layers are used in order to slow down λ-ageing and oxygen diffusion. Calculations of changes in the gas concentration in the pores of PU foams of long pre-insulated plastic pipes with inner and outer diffusion barrier layers are of practical interest [4–6].

In general, analytical solutions for the dynamic problem of radial diffusion/ heat conductivity of three-layer pipes (pressure pipe, heat insulation, protective layer) are complicated [7–9] and very difficult to use for numeral evaluation. If outer and inner surfaces of heat insulation have barrier layers, the spatial distribution of diffusion gas concentration is almost uniform [5, 10] and concentration changes over time can be described by a simple equation (see below). Heat conductivity of systems with negligibly small inner thermal resistance is described in [11].

Diffusion equation for isotropic medium [12]

$$\frac{\partial C}{\partial t} = div(D\,gradC)\,, \quad (1)$$

C – gas concentration, t – time, D – diffusion coefficient. Equation (1) is true for materials where chemical reactions are complete and/or no dissociation of the gas in question is present. Otherwise, the first part of the equation must contain a gas source function.

Assuming that far from the ends of long non-corrugated heat-insulated pipe with inner and outer barrier layers, the distribution of gas concentration in the heat insulating layer is near to uniform and gas diffusion happens only in radial direction, after integration of the equation (1) upon volume of thermal insulating layer within the given length, we derive:

$$\frac{\partial \overline{C}}{\partial t} = \frac{S_e}{V_f}\left(D_f\frac{\partial C}{\partial r}\right)_{r=R_e} - \frac{S_i}{V_f}\left(D_f\frac{\partial C}{\partial r}\right)_{r=R_i}$$

$$\left(\frac{S_{i,e}}{V_f} = \frac{2R_{i,e}}{R_e^2 - R_i^2}\right). \quad (2)$$

where $\overline{C}$, D_f – respectively, the concentration averaged over volume and efficient gas diffusion coefficient in the insulation material, V_f, S_i, S_e – volume of insulation and squares of its inner and outer surfaces in the given length, R_i и R_e – inner and outer radiuses of heat insulating layer. If gas diffusion is determined by barrier layers and its concentration slightly differs from the average value $\left(\left|C - \overline{C}\right| \ll \overline{C}\right)$, then, omitting the influence of pressure pipe and protective jacket, we get the following for thin barrier layers:

$$\left(D_f\frac{\partial C}{\partial r}\right)_{r=R_i} = D_i\frac{\overline{C} - C_i}{e_i}\,, \qquad \left(D_f\frac{\partial C}{\partial r}\right)_{r=R_e} = -D_e\frac{\overline{C} - C_e}{e_e}$$

$$(e_i \ll R_i,\ e_e \ll R_e)\,, \quad (3)$$

where D_i, D_e – gas diffusion coefficients through the materials of inner and outer barrier layers, e_i, e_e – thicknesses of these layers, C_i, C_e – gas concentration inside the pressure pipe and in the air surrounding the protective layer. The integration of equation (3) into equation (2) gives:

$$\frac{d\overline{C}}{dt} = -\left(\frac{1}{\tau_i} + \frac{1}{\tau_e}\right)\overline{C} + \frac{C_i}{\tau_i} + \frac{C_e}{\tau_e}\,.$$

$$\left(\tau_{i,e} = \frac{V_f e_{i,e}}{S_{i,e}D_{i,e}}\right). \quad (4)$$

If $C_{i,e}$ and $D_{i,e}$ are the values that don't depend on time, the differential equation (4) solution in compliance with initial condition $\overline{C}(0) = C_0,$, can be recorded as:

$$\overline{C} = (C_0 - C_s)e^{-\frac{t}{\tau}} + C_s$$

$$\left(\tau = \frac{\tau_i \tau_e}{\tau_i + \tau_e},\quad C_s = \frac{\tau_i C_e + \tau_e C_i}{\tau_i + \tau_e}\right), \quad (5)$$

where τ – concentration change characteristic time, C_s – stationary concentration (marginal value $\overline{C}$ at $t \to \infty$, in the reality at $t \gg \tau$). At $C_0 = C_s$ gas concentration in heat insulating layer remains constant $(\overline{C} = C_0 = const)$, as in this case at $C_i \neq C_e$ gas influx in PU foam through one barrier layer equals its deflux through the other. Equation (5) allows to calculate the average gas concentration value change in time in heat insulating layer made of PU foam between two barrier layers for different practical cases. For example, for the pipe in open air without heat medium we have $C_i = C_e = C_s$ (average mass content of basic gases in dry atmosphere is N_2 – 75,51%, O_2 – 23,14%, Ar

– 1,29%, CO_2 – 0,05% [13]). After determining concentration of the given gas in PU foam upon given time lapse of pipe storage at the warehouse, using equation (5), we can then use it as a starting value when calculating gas concentration during pipeline operation.

Diffusion coefficient of barrier layer materials can change in time due to materials ageing, impact of humidity and/or temperature change. If such changes in both layers during gas diffusion process can be described by the same time function $(D_{i,e} = D_{i0,e0} \, F(t)$, where D_{i0}, D_{e0} are constants), then it follows from equation (4) that the equation (5) is still valid if $D_{i,e}$ in the expressions for $\tau_{i,e}$ are replaced by the respective time-averaged values. At $C_i = C_e$ such replacement in equation (5) is true in case when the changes of diffusion coefficient values are described by different time functions $(D_{i,e} = D_{i,e}(t))$. Solution of differential equation (4) in quadratures can be written for the common case when diffusion coefficient of barrier layers materials and gas concentration inside and outside the pipe depend on time $(D_{i,e} = D_{i,e}(t), C_{i,e} = C_{i,e}(t))$.

The influence of pressure pipe and protective jacket on the radial gas diffusion in plastic pipes with heat insulation between barrier layers can be ignored when using following conditions:

$$\frac{e_p}{D_p} \ll \frac{e_i}{D_i}, \qquad \frac{e_c}{D_c} \ll \frac{e_e}{D_e}$$

$$(e_p \ll R_i, \; e_c \ll R_e). \quad (6)$$

where e_p, e_c – thicknesses, accordingly, of pressure pipe wall and protective jacket (usually $e_{i,e} \ll e_{p,c}$), D_p, D_c – their diffusion coefficients. Spatial distribution of the gas in the PU foam layer is near to uniform if the following condition is followed:

$$\frac{e_f}{D_f} \ll \frac{e_{i,e}}{D_{i,e}},$$

where e_f – thickness of the heat insulation layer. Temporal variation of gas concentration averaged over a volume of thermal insulation layer in such pipes is described by equation (5), and concentration changes in the thickness of the pressure pipe ΔC_p and of the jacket ΔC_c are low and equal

$$\Delta C_{p,c} = \left| \overline{C} - C_{i,e} \right| e_{p,c} D_{i,e} / \left(e_{i,e} D_{p,c} \right).$$

For example, let's calculate the characteristic time of oxygen concentration change in PU foam for the pipes stored at the warehouse with relatively high diffusion coefficient value through the barrier layers $D_i = D_e = 7 \cdot 10^{-16}$ m²/s and thicknesses of barrier layers $e_i = 0,1$ mm и $e_e = 0,01$ mm. Then, e.g., for the pipes with sizes 32/63, 75/125, 140/225, 160/270 (V_f/S_i = 19, 32, 50, 82 mm, V_f/S_e = 10, 20, 32 and 45 mm accordingly) we get $\tau \approx \tau_e$ = 4,5, 8,9, 14,5 and 20,4 years ($\tau_i \gg \tau_e$). Graphs of normalised concentration value changes in time

$$\left(\overline{C} - C_0 \right) / \left(C_s - C_0 \right) \approx 1 - exp(-t/\tau_e)$$

(here $C_s = C_i = C_e$ и $C_0 < C_s$) for these values τ_e are shown on the picture.
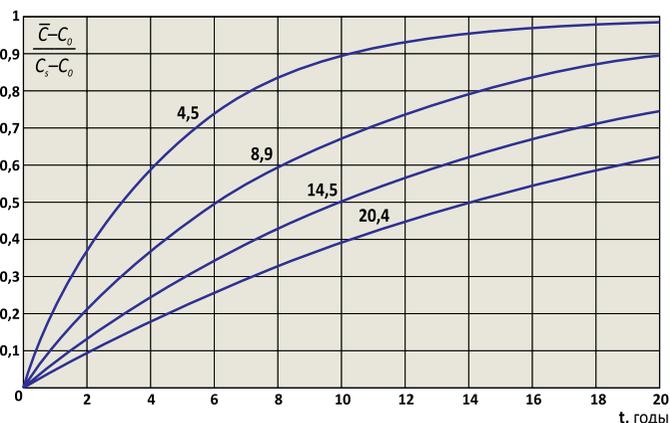
Increase of the normalised oxygen concentration in the PU foam in time for the pipe sizes 32/63, 75/125, 140/225, 160/270 with barrier layers of various thickness e_i = 0.1 mm, e_e = 0.01 mm and the same diffusion coefficient $D_i = D_e =$ 7·10¹⁶ m²/s (values $\tau \approx \tau_e$ = 4.5, 8.9, 14.5 and 20.4 years correspond to relations values V_f/S_e = 10, 20, 32 and 45 mm).

In the given situation of relatively high permeability of the barrier layers and thin jacket characteristic time of oxygen concentration increase in PU foam for the pipes with a small relation V_f/S_e is not great, and after storing them for several years there will be significant increase of concentration (time of the normalised value increase up to half of the maximum is $t = 0,69\tau$).

To maintain the initial concentration, we obviously need to increase the thickness of the outer barrier layer for such pipes and/or use barrier films with lower oxygen permeability coefficient. The thickness increase of the outer barrier layer to the thickness of the inner layer $e_i = e_e = 0,1$ mm for the pipes with V_f/S_i = 19 mm, V_f/S_e = 10 mm will give τ_e = 45.3 years and, considering τ_i = 86.1 years, we get τ = 29.7 years. The above shown diffusion coefficient matches type G of EVAL™ gas barrier films (48% of ethylene) at 20°C and relative humidity of 65%, having the highest oxygen permeability out of grades presented in [14]. The type L films (27% of ethylene) oxygen permeability at the same conditions is 15 times lower and corresponding value of τ_e for the same pipe in the earlier given example (e_i = 0.1 mm, e_e = 0.01 mm) exceeds 68 years.

Equation (5) is true in more general cases when conditions (6) are not fulfilled and there is a need to include the influence of pressure pipe and protective jacket. In these cases, we have to change τ_i and τ_e in this solution, accordingly, to $\tau_i + \tau_p$ и $\tau_e + \tau_c$, where $\tau_{p,c} \approx V_{f} e_{p,c}/(S_{i,e} D_{p,c})$ (we assume that $e_i \ll e_p \ll R_i$, $e_e \ll e_c \ll R_e$). Gas concentration change values in these layers equal

$$\Delta C_{p,c} = \left|\overline{C} - C_{i,e}\right|/(1 + \tau_{i,e}/\tau_{p,c})$$

(the influence of adhesion layers to diffusion process in reinforced plastic pipes can be counted using the similar method). In quasi-stationary mode at $\overline{C} \approx C_s$ for the concentration change in the pressure pipe wall we get

$$\Delta Cp \approx |\,C_e - C_i\,|\,\tau_p\,/(\tau_i + \tau_p + \tau_e + \tau_c).$$

This equation shows that at low oxygen concentration inside the pressure pipe ($C_i \ll C_e$) and $\tau_{i,e} \gg \tau_{p,c}$ maximal concentration on its outer surface $\Delta C_p \approx C_e\,\tau_p\,/(\tau_i + \tau_e)$ is much lower than the concentration on the outer surface of the protective jacket. For the heat insulated metal pipes with outer barrier layer we have $\tau = \tau_e$, $C_s = C_e$ ($\tau_p \rightarrow \infty$ at $D_p \rightarrow 0$), and solution (5) in this case is the same as the solution for vapour partial pressure from [10]. The given method for calculation of gas concentration in PU foam heat insulation of plastic pipes with barrier layers can be used to determine their shelf life when initial and marginal conditions are known and for calculation of concentration at pipeline operation.

References:

1. Larsen C.T., Togeskov P., Leuteritz A. Extending the Service Life of Pre-Insulated Pipes – Analyses of Diffusion Rates Through PE and Impact on Ageing. EuroHeat & Power Int., 6, 11, 2009.
2. Persson C., Reidhav C., Jarfelt U., Ramnäs O. Insulating Performance of Flexible District Heating Pipes. 10-th Int. Symp. on District Heating and Cooling, 2006.
3. Krecielewska E., Menard D. Thermal Conductivity Coefficient of PUR Insulation Material from Pre-Insulated Pipes after Real Operation on District Heating Networks and After Artificial Ageing Process in Heat Chamber, 14-th Int. Symp. on District Heating and Cooling, 2014.
4. Brodt K.H., Brood R.R.H., Bart G.C.J. Permeation of Gas Barrier Layers, J. Thermal Insul. and Bldg. Envs., 19, 1995.
5. Ivanov V., Karpukhin O., Ivanov A., Ermilova A. Characteristics of oxygen diffusion in multi-layered plastic pipes. – Plastic pipes, Issue 1, 2015.
6. Patent EP 0634602 A1, Thermally insulated pipe, 1995.
7. Lu X., Terlova P., Viljanen M. An efficient analytical solution to transient heat conduction in a one-dimensional hollow composite cylinder, J. Phys. A: Math. Gen., 38, 2005.
8. Monde M., Arima H., Liu W., Mitutake Y., Hammad J.A. An analytical solution for two-dimensional inverse heat conduction problems using Laplace transform, Int. J. Heat Mass Transfer, 46, 12, 2003.
9. Tugolukov E.N. Heat conductivity problem solving using finishing integral transformation method for automated design of machinery for chemical industry, Tambov: TGPU, 2006.
10. Svanström M., Ramnäs O., Olssen M., Jarfelt U. Mass Transfer of Carbon Dioxide through the Polyethylene Casing of District Heating Pipes, J. Building Phys., 21, 1997.
11. Kreith F., Black W. Basics heat transfer, Moscow, Mir 1983.
12. Crank J. The Mathematics of Diffusion, Oxford: Clarendon Press, 1975.
13. Zaozerskiy N.N., Kotlyarov P.B., Platonov F.P. and others. Inorganic chemistry, Moscow, Vysshaya Shkola, 1965.
14. EVAL™ – Kuraray specification booklet.