

ANNA MUSZ¹, MARCIN K. WIDOMSKI¹, BEATA KOWALSKA¹

BENZENE PROPAGATION DURING TURBULENT FLOW IN PEHD WATER SUPPLY PIPES

The results of laboratory and numerical studies of benzene propagation migrating from PEHD pipe to water in turbulent flow conditions have been presented. Laboratory studies were performed on 30 meter long closed-loop installation consisting of PEHD 80 32×3.0 mm polyethylene pipes during the 168 h of experiment. Measurements of benzene concentration in water were conducted by the gas chromatography-mass spectroscopy (GC-MS) method. Numerical calculations of benzene propagation in water were performed with the Fluent, Ansys Inc. Our studies showed a clear increase of benzene concentration in water resulting from its migration to water from the pipe material. The developed numerical model was positively validated.

1. INTRODUCTION

Polyethylene is nowadays commonly used in production of PEHD pressured pipes applied to construction of water supply systems, domestic water installations and to rehabilitation of the old water pipelines. Water supply pipes made of PEHD are endangered by influence of the external conditions, both, during the production and later exploitation in civil and environmental engineering. Defects and degradation of the material of plastic pipes and release of organic compounds from pipe material to water body may result from UV radiation, increased temperature, mechanical loads and influence of disinfectants applied to drinking water [1, 2]. Organic compounds migrating to water may cause changes in its organoleptic properties, i.e. deterioration of its taste and smell. The adverse changes in pipe material triggered by external atmospheric conditions usually start at the outer surface of a pipe and progress to its inner surface, while deterioration caused by influence of oxidizing compounds contained in water body, in turn, proceeds from the inner to the external surface of the pipe [2].

¹Lublin University of Technology, Faculty of Environmental Engineering, ul. Nadbystrzycka 40B, 20-618 Lublin, Poland, corresponding author M.K. Widomski, e-mail: M.Widomski@wis.pol.lublin.pl

Available literature reports [3–5] show that the migration of additional compounds applied to polymers during their production as well as the products of their degradation is triggered by damages of polymer materials. Chemicals released from PE pipes to water were categorized by Denberg et al. [1] as: i) antioxidants, ii) products of antioxidants degradation, iii) decollated PE chains with functional, polar oxygen group.

Moreover, it was shown that the dominant species released to water and influencing its properties are antioxidants: Irganox 1010, Irganox 1035, Irganox 1076, 2,6-di-*tert*-butyl-4-methylphenol (BHT), products of degradation of antioxidants (4-ethyl phenol, 4-*t*-butyl phenol, 2,6-di-*t*-butyl-*p*-benzoquinone, 2,4-di-*t*-butyl phenol, 3,5-di-*t*-butyl-4-hydroxy styrene, 3,5-di-*t*-butyl-4-acetophenone, 3,5-di-*t*-butyl-4-hydroksy acetophenone) and VOCs: aromatic (e.g. benzene), toluene, naphthalene, 1,3,5-trimethyl benzene, esters, ketones and aldehydes [1, 3, 4]. The research conducted by e.g. Skejvark et al. [4] showed that the danger of benzene migration to water body is highly possible.

Benzene, aromatic hydrocarbon is one of the substrates commonly applied to production of PEHD pipes, and, according to the dose, it is considered a toxic substance, which can be absorbed by the skin, respiratory system and gastrointestinal tract of humans, strongly irritating for skin and pulmonary airways and, finally, demonstrating carcinogenic capabilities [6]. Its adverse influence on the central nervous and circulatory systems was also proved, the reduction of white cells number, resulting in decrease of humans body immunity, as well as in number of blood cells were reported [7].

Requirements of 98/83/EC UE Directive and several international and national standards, including Polish requirements [8, 9], where the maximum allowable concentration of benzene in drinking water was presented as $1 \mu\text{g}\cdot\text{dm}^{-3}$, lead to the necessity of new methods for monitoring changes of water quality occurring during water distribution, including measurements of organic compounds originated in pipe material [6, 10].

Thus, the increasing popularity of numerical modeling changes of water quality inside water supply pipelines is reasonable. Such modeling allows the analysis of migration, propagation and decay of polluting compounds, supporting proper designing and management of water distribution networks [11–15].

Application of computational fluid dynamics (CFD) to numerical modeling of water quality may enable the calculation of pollutants propagation in water supply pipelines in various dynamic flow conditions [1, 16]. One of the most popular commercial CFD software of proven computational capabilities is Fluent, Ansys Inc. The mathematical description of fluid dynamics applied to Fluent software is based on well known, principal in CFD, mass, momentum, angular momentum and fluid flow (Navier–Stokes) equations [17]. These equations, supported with properly assumed initial and boundary conditions, allow effective modeling of fluid motion (also multiphase fluids) under laminar or turbulent flow conditions, for steady or transient flows. Qualitative calculations covering transport, mixing, reactions and decay of selected species are based on mass conservation equations for a given compound [18]:

$$\frac{\partial}{\partial t}(\rho Y_i) + \nabla(\rho \mathbf{u} Y_i) = -\nabla \mathbf{j}_i + R_i + S_i \quad (1)$$

where: Y_i – local mass fraction of i species, \mathbf{j}_i – vector of diffusion flux of i species, R_i – production rate of i species by chemical reaction, S_i – creation rate of i species by addition from dispersed phase or any other possible source, \mathbf{u} – flow velocity vector.

The aim of this paper is to present the results of our laboratory measurements and numerical modelling of benzene propagation inside water body circulating with a constant rate during turbulent flow through a closed loop made of PEHD pipes. Numerical calculations of benzene concentrations inside water stream were performed with the Fluent, Ansys Inc. and empirically validated by comparison to the results of laboratory measurements.

2. MATERIALS AND METHODS

The PEHD pipes selected to the studies were produced according to the Polish national standard PN-EN 12201-2: 2012 [19]. The concentration of benzene was monitored in the laboratory installation specially constructed for this purpose (Fig. 1).

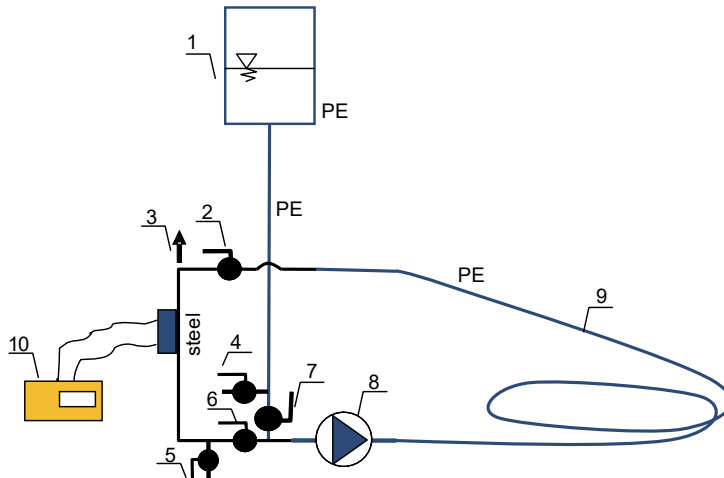


Fig. 1. Scheme of laboratory installation: 1 – deionized water tank, 2 – valve, 3 – air relief valve, 4 – relief valve, 5 – sampling point, 6 – regulatory valve, 7 – inlet valve, 8 – circulation pump, 9 – plastic pipe PEHD 32.0×3.0 mm, L=30.0 m, 10 – flow meter PortaFlow 300

A 30 m long laboratory installation consisted of new PEHD 80 pipes of the diameter 32×3.0 mm purchased directly from the producer. The inner area of the pipe wall was 2.45 m² while the volume of water body inside the closed loop was equal to 15.9 dm³.

Before the experiment, the pipes were flushed with deionized water prepared by Milli-Q (Millipore, Molsheim, Germany) with at least three exchanges of water inside the laboratory installation. Next, the pipe loop was filled with deionized testing water. Deionized water used in flushing and the main tests was characterized by $\text{TOC} \leq 0.5 \mu\text{g}\cdot\text{dm}^{-3}$ and resistivity of $18.2 \text{ M}\Omega\cdot\text{cm}$. The constant rate of water flow $v = 2.53 \text{ m}\cdot\text{s}^{-1}$ ($Re = 54\,816$) assuring turbulent flow was sustained with a circulation pump. Sampling was performed for the following seven days, in the following time steps: 0, 2, 4, 6, 8, 12, 24, 48, 72, 96, 120, 144 and 168 h. The adopted sampling timing resulted from literature studies and was reflecting water age in water supply lines [20, 21]. After each sampling, water inside the laboratory installation was supplemented with ca. 200 cm^3 of fresh deionized water. Water samples were collected in 40 cm^3 glass containers and kept at the temperature below $4 \text{ }^\circ\text{C}$ before being analyzed.

Measurement of benzene contents. The measurements were performed by the GC-MS method, being a combination of gas chromatography and mass spectroscopy, two sophisticated instrumental techniques used to analyse organic compounds by the most effective method of identification and measurement of trace amounts of VOCs in complex natural matrices. This method is used to measure the contents of organic compounds in biological materials, food, medicines, water and air [22, 23].

Water samples for the measurement were placed in Supelco 20 cm^3 conical glass containers. The samples were heated in water bath at $35 \text{ }^\circ\text{C}$ for 30 min, then extracted to the solid phase using SPME (SUPELCO $100 \mu\text{m}$ polydimethylsiloxane) fibre for 15 min at $25 \text{ }^\circ\text{C}$. The prepared sample of extract was dosed directly from the fibre to the dispenser of a gas chromatograph Trace-Ultra/Polaris Q (Thermo, USA) combined with a mass spectrometer Polaris Q, type Ion Trap. Operational conditions of the chromatograph unit were as follows: temperature of the dispenser operation – $250 \text{ }^\circ\text{C}$ in splitless option during 2 min; temperature of ion source – $250 \text{ }^\circ\text{C}$; temperature of detector's transfer line – $250 \text{ }^\circ\text{C}$, temperatures of column operation: – start $45 \text{ }^\circ\text{C}$, isotherm 2 min, increase $16 \text{ }^\circ\text{C}\cdot\text{min}^{-1}$ until $250 \text{ }^\circ\text{C}$ – isotherm 2 min. Helium 99.9996% was used as a carrier gas in mobile phase with the constant rate flow of $1.2 \text{ cm}^3\cdot\text{min}^{-1}$. The mass spectra Full Scan: 42 – 150 were obtained as measurement results. The time of benzene retention was equal to 5.29 min. The quantitative benzene analysis was performed by measurement of the surface area of the bands corresponding to characteristic ions (Base peak m/z – 51, 77, 78).

Detection limit of benzene concentration in tested water samples was $0.02 \mu\text{g}\cdot\text{dm}^{-3}$, while the quantification limit (QL) was $0.06 \mu\text{g}\cdot\text{dm}^{-3}$.

Numerical modeling. A 3D numerical model of benzene migration and propagation inside stream of water flowing with a constant rate through the closed loop pipeline was developed in the commercial software Fluent 6.3, Ansys 14.0, Ansys Inc. The developed model, consisting of 179 090 nodes and 149 783 elements reflected the closed-loop

made of PEHD pipe of the diameter of 32×3.0 mm and 30 m long (Fig. 2). The presented modeling attempt required requisite simplifications. The developed model did not contain any circulation pump – the modeled water flow with a constant mean rate was obtained by assigning the constant axial rate of flow to the limited control volume [24, 25]. Then, the momentum of a plug flow from the control volume was transferred to the main domain of the modeled water body, in which a typical parabolic distribution of flow rate was afterwards developed.

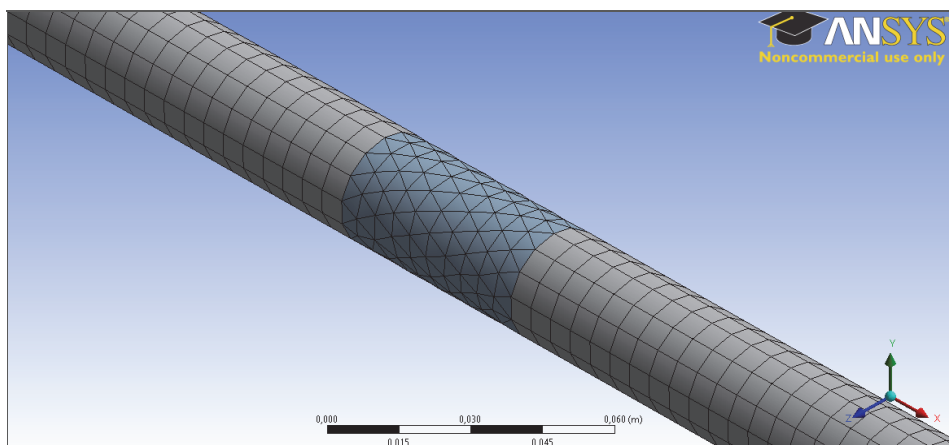


Fig. 2. Selected part of the developed numerical model with a visible region of plug flow

Benzene decay and chemical reactions occurring inside the modeled water body between benzene and other species available in water were neglected. We also neglected the changes of benzene concentration caused by water sampling for chemical analyses and addition of pure distilled water in order to maintain the total volume of water body.

The assumed duration of simulation covered 168 h of water flow with a constant mean flow rate equal to $2.53 \text{ m} \cdot \text{s}^{-1}$, reflecting the mean flow rate of water in our laboratory measurements. The performed numerical calculations of water flow were based on the standard k - ε model of turbulence for incompressible fluid [26, 27]. The parameters of liquids used in simulation are presented in Table 1.

Table 1

Characteristics of liquids used in numerical modeling

Material	Molar weight [g·mol ⁻¹]	Temperature [K]	Density [kg·m ⁻³]	Dynamic viscosity [kg·m ⁻¹ ·s ⁻¹]
Water	18.02	299	998.2	$8.68 \cdot 10^{-4}$
Benzene	78.11	–	875.0	–

The inner roughness for PEHD was assumed according to the information supplied by the producer as 10^{-6} m. The necessary diffusion coefficient for benzene was accepted according to the available literature reports as 10^{-15} $\text{m}^2 \cdot \text{s}^{-1}$ [1, 5]. Benzene mass fraction at $t = 0$ was assumed as 1.29×10^{-10} reflecting the pollutant concentration equal to 1.29×10^{-4} $\text{g} \cdot \text{m}^{-3}$, according to the results of our laboratory studies. The first type of boundary condition, the Dirichlet condition, assuming the time constant concentration of benzene at the outer boundary of modeled water body domain, in the boundary layer, located close to the pipe's inner wall surface, calculated by the trial and error method according to the results of our laboratory studies, was accepted to our simulation. We assumed the benzene mass fraction of 4.03×10^{-9} , reflecting the concentration 8.8×10^{-3} $\text{g} \cdot \text{m}^{-3}$ as the boundary condition. Our model calculations were performed with a constant time step equal to 60 s. The results of simulations covered 3D distributions of the flow rate, axial and radial rates, turbulence energy and intensity as well as mass fraction of benzene in water.

Model validation was performed based on the root mean square error (RMSE), normalized root mean square error – normalized to the range of data (see Eq. (2)), the correlation coefficient and the Nash-Sutcliffe model efficiency coefficient (Eq. 3).

The dimensionless normalized root mean square error was calculated according to the formula:

$$\text{NRMSE} = \frac{\text{RMSE}}{X_{\text{obs, max}} - X_{\text{obs, min}}} \quad (2)$$

where RMSE is the root mean square error, $X_{\text{obs, max}}$ is the maximal observed value and $X_{\text{obs, min}}$ – the minimal observed value.

The Nash–Sutcliffe model efficiency coefficient was calculated from to the equation:

$$E = 1 - \frac{\sum_{i=1}^n (X_{\text{obs, } i} - X_{\text{model, } i})^2}{\sum_{i=1}^n (X_{\text{obs, } i} - X_{\text{obs, mean}})^2} \quad (3)$$

where $X_{\text{obs, } i}$ is the observed value and $X_{\text{model, } i}$ is the modelled value at time/place i , $X_{\text{obs, mean}}$ is the mean observed value.

The Nash–Sutcliffe coefficient can take values starting from 1, meaning the perfect match of the model to the observed values, through 0, showing that the model predictions are as accurate as the mean of the observed data, to $-\infty$. Thus, negative values of E show that the observed mean is a better predictor than the modeling results.

3. RESULTS AND DISCUSSION

3.1. RESULTS OF LABORATORY MEASUREMENTS

The observed time dependent concentration of benzene in water body measured in water sampled from the laboratory installation is presented in Table 2.

Table 2

Measured benzene concentration in water

Time duration, h	0	2	4	6	8	12	24	48	72	96	120	144	168
Benzene concentration $\times 10^3$, $\text{g}\cdot\text{m}^{-3}$	0.13	0.25	0.33	0.68	0.75	0.82	0.88	1.17	1.21	1.23	1.50	1.62	1.93
Temperature, $^{\circ}\text{C}$	25.0	24.5	26.0	26.0	26.0	26.0	27.0	26.5	27.5	27.0	27.0	27.0	27.0

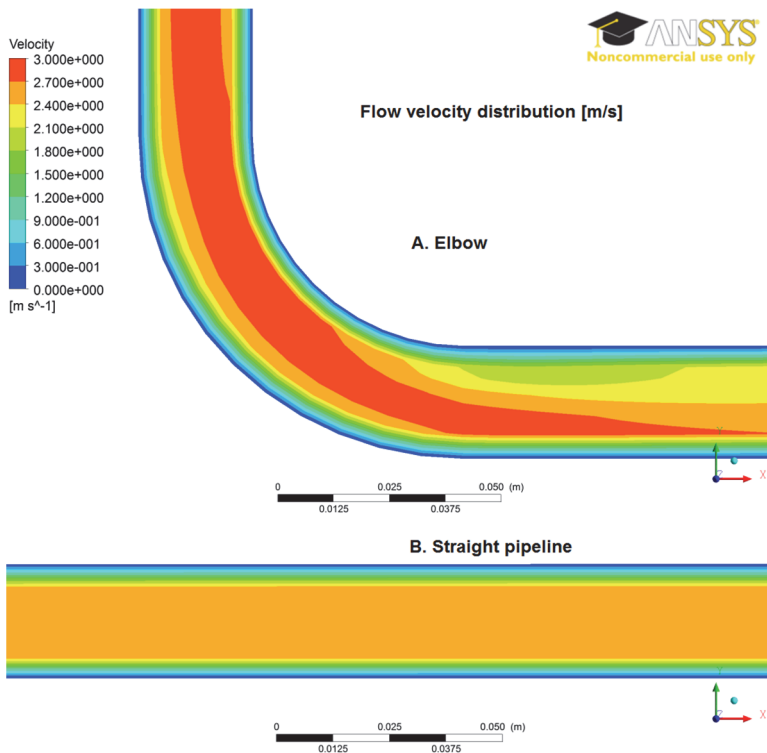


Fig. 3. Fluid flow velocity distribution for selected parts of the modeled pipeline

During the first hours of the experiment, for ca. 24 h, a rapid increase of benzene concentration was observed (the increase of $7.5 \times 10^{-4} \text{ g}\cdot\text{m}^{-3}$ was noted, from $1.3 \times 10^{-4} \text{ g}\cdot\text{m}^{-3}$ to $8.8 \times 10^{-4} \text{ g}\cdot\text{m}^{-3}$), then the increase ratio was smaller and finally it was equal to $1.93 \times 10^{-3} \text{ g}\cdot\text{m}^{-3}$ after 168 h of the experiment. The relative increase between 24th and

168th hour of the experiment was $1.05 \times 10^{-3} \text{ g} \cdot \text{m}^{-3}$. The maximum observed benzene concentration after 168 h is nearly two-fold higher than that allowed by the Polish standards for drinking tap water ($1 \mu\text{g} \cdot \text{dm}^{-3}$, [9]). The noted presence of benzene, above the QL of the applied method, at the time of the first sampling assumed as $t = 0$, results probably from time duration required to filling the installation and air removal from the pipelines, during which water was in constant contact with the pipe material.

Application of the circulation pump (Fig. 1) allowing water flow through the closed-loop pipeline with a constant flow rate resulted in minor increase of water temperature (Table 2). Its maximal increase was $3.0 \text{ }^\circ\text{C}$ with respect to the minimal value measured after 2 h of experiment. In order to ensure a nearly constant level of water temperature, the tested pipeline was located in a tank at $12 \text{ }^\circ\text{C}$.

3.2. RESULTS OF NUMERICAL CALCULATIONS

The results of numerical calculations used in our analyses of benzene distribution in the flowing water body covered distributions and mean values of benzene mass fraction, directly connected to the hydraulic characteristics of the water stream. The water flow rate for the selected part of the modeled domain is shown in Fig. 3.

The exemplary results showing the benzene distribution in water flowing through the PEHD closed loop pipeline with a constant flow rate (mean value $2.53 \text{ m} \cdot \text{s}^{-1}$), after 6, 24, 48, 96 and 168 h are presented in Fig. 4.

A close relation between hydraulic characteristics of flow, i.e. distribution of flow rate (Fig. 3) and the distance from the inner surface of pipe wall and mass fraction of modeled pollutant is clearly visible in Fig. 4. The higher values of modeled concentration of benzene were observed in the close proximity of pipe wall, in the boundary layer of flowing stream where the flow rate is limited, even to zero, while the lowest pollutant content was noted in the main part of the water stream, close to the axis of the pipeline, in the zone of the highest flow rate.

Full mixing of the modeled pollutant inside the water body was observed after 168 h of simulation, which is visible in Fig. 4, reflecting the distribution of benzene mass fraction after the final step of simulation.

Figure 5 presents the calculated and measured values of benzene concentration in water body filling the previously described laboratory installation.

The difference between the final calculated and measured benzene concentration was equal to 1%. The shape of curve reflecting the time varied increase of benzene concentration in water, originated in migration of the tested compound from the material of pipe wall, is similar to curves reported in literature for other organic compounds released from the pipes material to water in the dynamic conditions of flow [24, 25, 28].

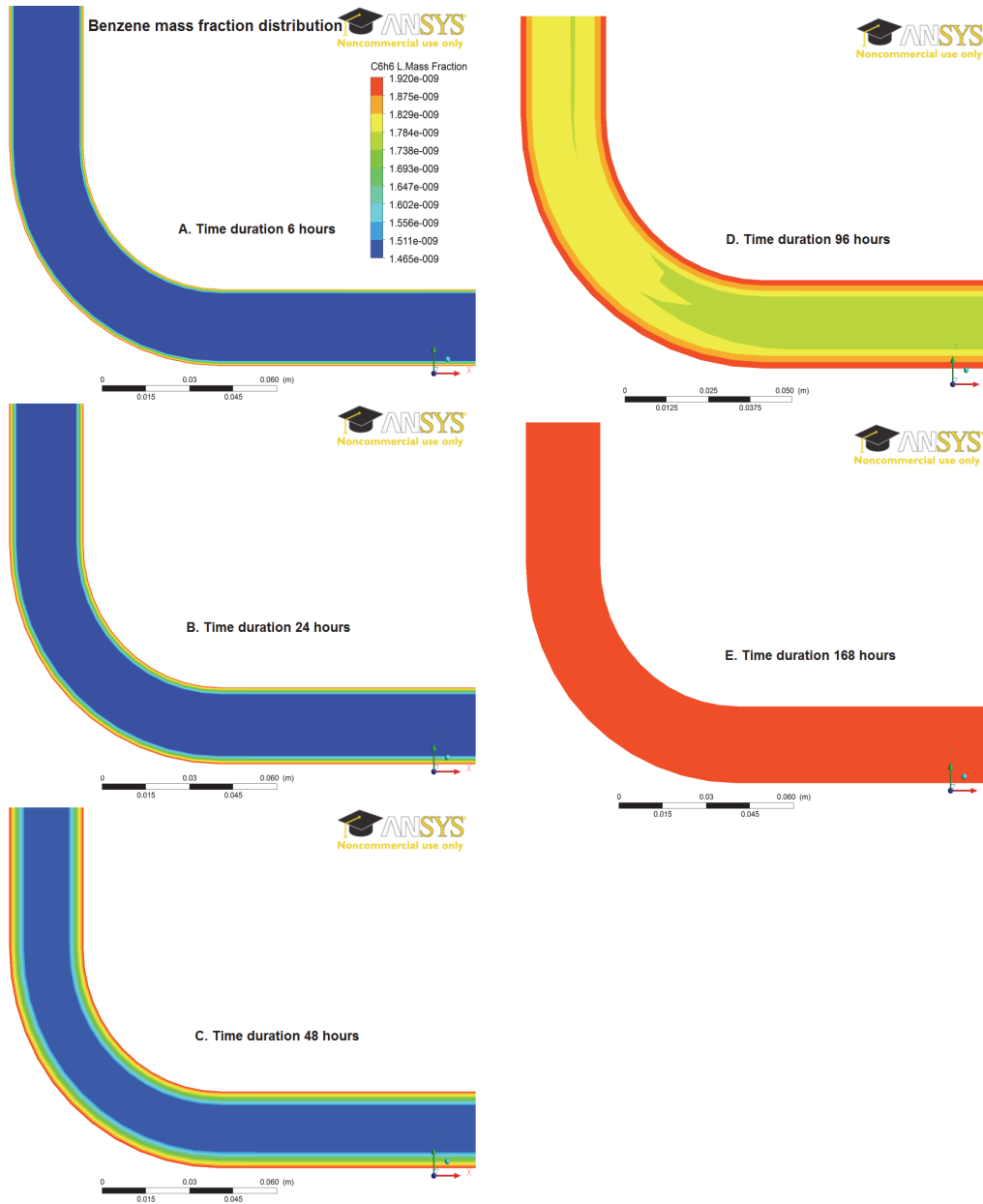


Fig. 4. Benzene mass fraction distribution for the selected part of the modeled pipeline and various time durations: 6, 24, 48, 96 and 168 h

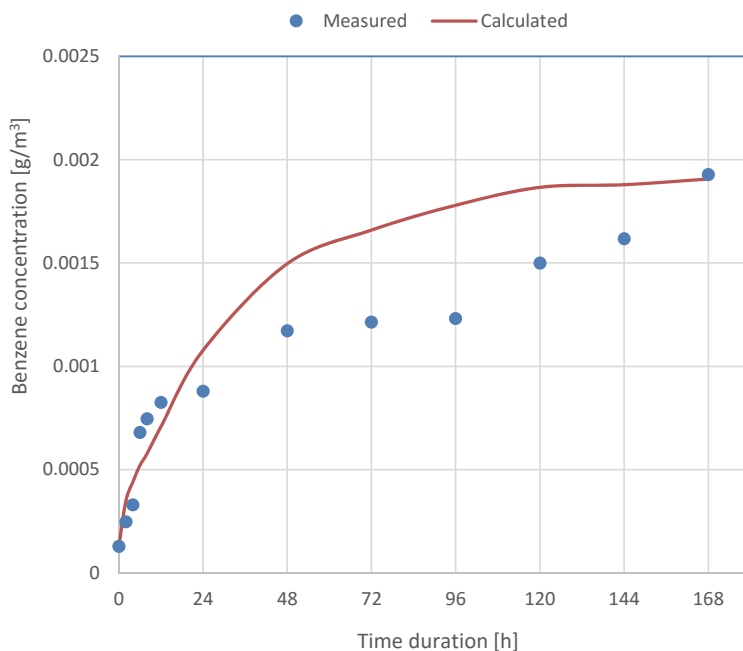


Fig. 5. Calculated and observed time dependences of the benzene concentration in water

The empirical validation of the developed model, performed by comparison of measured and calculated values of benzene concentration showed satisfactory agreement, despite the fact that results of our calculations are higher than the measured values, especially in the range 24–144 h. The calculated RMSE was equal to $2.69 \times 10^{-4} \text{ g}\cdot\text{m}^{-3}$, while NRMSE was at the level of 0.151. The values of $R = 0.9478$ and $R^2 = 0.88$ for $P = 0.05$ were obtained. Finally, the calculated Nash–Sutcliffe coefficient was 0.74, which shows quite good agreement between modeled and observed values of benzene concentration.

4. SUMMARY AND CONCLUSIONS

The laboratory measurements of benzene concentration in water performed during migration test in dynamic, turbulent conditions for the mean flow rate of $2.53 \text{ m}\cdot\text{s}^{-1}$ showed remarkable migration of the tested compound resulting in a clear increase of its concentration in water body. The observed relative increase of benzene concentration in water after 168 h of the laboratory experiment was ca. tenfold, which may be connected to increased temperature of water (standard temperature of water in tap installation is usually below $20 \text{ }^\circ\text{C}$, in the range $12\text{--}18 \text{ }^\circ\text{C}$) resultant from high friction forces in turbulent flow, cooling of circulation pump, indoor conditions and time duration of the experiment. The maximal measured value of benzene concentration was $1.93 \times 10^{-3} \text{ g}\cdot\text{m}^{-3}$. No literature

data is available on changes of benzene concentration in water during turbulent flow through PEHD pipelines. The concentration of ca. $1 \mu\text{g}\cdot\text{dm}^{-3}$ was measured during the standard, static migration test. Results of our numerical studies show close relation between distribution of flow rate and mass fraction of benzene in water body. The highest benzene content was observed in the location of the lowest flow rate. The measured and calculated shape of curves representing time dependent increase of benzene concentration in water due to migration of the pollutant from the material of pipe wall during turbulent flow, resembles the shape of curves reported in the literature for the different species migrating from plastic pipes to water.

The obtained satisfactory agreement between measured and calculated values of benzene concentration in water shows that application of Fluent, and generally CFD modeling, may be a successful tool in studies focused on migration of pollutants from pipe material and their propagation inside water body during turbulent flow.

The presented laboratory and modeling studies should be, in our opinion, continued, first of all, introducing more sophisticated, time dependent Dirichlet and Neumann type boundary conditions, and considering reactions of benzene with the other compounds present in water to a developed numerical model in order to obtain the better match between the observations and results of modeling. Then, our model should be extended to cover various flow rates, including laminar flow to allow the assessment of influence of hydraulic conditions of benzene migration rate and distribution.

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