TRACER DISPERSION – EXPERIMENT AND CFD

Rudolf Žitný
Czech Technical University in Prague, Czech republic

Abstract: Description of tracer distribution by means of dispersion models is a method successfully used in process engineering for fifty years. Application of dispersion models in reactor engineering, for characterization of flows in column apparatuses, heat exchangers, etc is summarized and experimental tracer techniques as well as CFD methods for dispersion coefficients evaluation are discussed. Possible extensions of thermal axial dispersion model (ADM) and a core-wall ADM model suitable for description of tracer dispersion in laminar flows are suggested as well as CFD implementation as 1D finite elements.

1. INTRODUCTION – 50 years anniversary of ADM

History of engineering applications of tracers is long, probably the first one concerns measurement of velocities and flow-rates in large water mains. Allen and Taylor (1923) measured flowrate by injecting a packet of salt into the pipe at one point and recorded electrical conductivity at a point downstream (diameter of pipe 1m, length 100m, mean velocity 1 m/s, $Re=10^6$, which means highly turbulent flow). They found that the time corresponding to the instant at which recorded conductivity was a maximum is a mean residence time and can be used for calculation of mean velocity. Similar conclusions have been achieved using different tracers, for example Hull & Kent (1952) injected radioactive tracer into a crude oil pipe nearly 300 km long and observed symmetrical nearly Gaussian responses (again in turbulent flows, Re=24000). However, this simple method of flowrate measurement fails, when applied to short pipes or laminar flows. The situation changed fifty years ago thanks to the pioneering work of Taylor[1] (1953, 1954) who formulated and experimentally verified the axial dispersion model (ADM) which has become popular in process engineering not only for its ability to describe an inert tracer dispersion in laminar/turbulent flows in pipes but also for modelling tubular reactors, column apparatuses, packed, bubble and fluidized beds, heat exchangers, extruders and similar apparatuses. Excellent books by Wen, Fan (1975), Nauman (1983), or Levenspiel (1989) have promoted dissemination of Taylor's concept to the chemical engineering community. Just looking at the list of papers presented in the respected journals on chemical engineering during last 3 years indicates that ADM concept is still vivid no matter that the CFD programs seem to be suitable for modelling most of the mentioned cases. In fact some of the CFD solutions are interpreted either in terms ADM or by using an equivalent compartment model (hybrid models). The reason is obvious: it is much more easier to calculate complicated chemical reactions (for example 3000 elementary reactions describing NOx production in a furnace, Faravelli et al (2001)) using a compartment model of several (ten) ideally mixed reactors than using hundred of thousands cells in CFD. And above all: the axial dispersion coefficient is an integral numerical characteristic enabling to compare different designs and evaluate performance of apparatuses.

[1] Sir Geoffrey Ingram Taylor (1886-1975) retired at 1952, but at the same time as he worked out the ADM model for laminar (1953), and turbulent (1954) flows, he invented a method for bulk viscosity measurement. Later on he studied movement through porous surfaces and the dynamics of sheets of liquids (last paper was published when Taylor was 83 years old, remarkable!). G.Taylor is of course better known by his earlier works on aerodynamic, turbulence, Taylor's vortices instabilities, motion of bubbles (his relationship for velocity of bubbles is still used in CFD modeling of bubble columns).

2. ADM: ANALYTICAL SOLUTIONS, CFD AND EXPERIMENTS

Taylor's experiments were simple: he observed motion of tracer (water solution of potassium permanganate) in a glass capillary (1 mm diameter) and evaluated mean concentration of tracer at different times and positions along a tube by colorometric method. Very small velocity (of the order mm/s) ensured fully developed laminar flow, when the tracer distribution is described by equation

$$\frac{\partial c}{\partial t} + 2u(1-r^2)\frac{\partial c}{\partial x} = D\left(\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial c}{\partial r}\right) + \frac{\partial^2 c}{\partial x^2}\right). \qquad (1)$$

where r is a dimensionless radial coordinate ($r=r^*/R$) and D is coefficient of molecular diffusion. Taylor transformed this equation to a floating coordinate system moving with the mean velocity u in the flow direction ($x \rightarrow \zeta = x-ut$) and considered situation at a sufficiently long time after the tracer injection (this time corresponds to diffusion transport across the radius $R=\sqrt{(\pi Dt)}$). Then he estimated radial concentration profile at $\zeta=const$ from Eq.(1) by neglecting axial molecular diffusion and assuming that $\partial c/\partial\zeta$ is independent of r (this crucial simplification has been questioned later by Taylor (1954b) himself and others, but proved to be correct for long time solution). Using this radial profile of concentration and parabolic velocity profile the net flow of tracer across the cross section $\zeta=const$ can be calculated and as a result linear relationship between $\partial c/\partial\zeta$ and mass flux is obtained with the axial dispersion coefficient D_e substituting diffusion coefficient D

$$D_e = \frac{R^2u^2}{48D}. \qquad (2)$$

Returning back to the fixed coordinate system the Eq.(1) reduces to the ADM

$$\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x} = D_e\frac{\partial^2 c}{\partial x^2}. \qquad (3)$$

This result can be interpreted as follows: Axial dispersion depends upon the velocity profile and radial mixing (this conclusion holds generally, for example in turbulent flows as well). Knowing velocity profiles the axial dispersion coefficient can be therefore derived in the same way for different cross sections (rectangular, elliptical), for non Newtonian liquids (power law and Bingham liquids), and even for packed beds of porous adsorbing particles, see Levenspiel (1989).

Distribution of tracer as an exact solution of Eq.(1) using series expansion was presented by Shankar and Lenhoff (1989) and asymptotic solutions, valid either for short or long time, were derived e.g. by Hunt (1977) and Vrentas & Vrentas (1988), (2000). Hunt (1977) derived asymptotic solution for short times making use of suitable transformation of Eq.(1) magnifying regions of front and rear wave of tracer formed by convection of initially planar layer. In these regions distribution of tracer is described by erf-functions (which could have been expected, because erf function describes the concentration response to a sudden step in 1D problem). Very interesting is the last paper Vrentas& Vrentas (2000), starting from the Eq.(1) transformed to the moving coordinate system, in the same way as Taylor did. Asymptotic solution for very short times was obtained by neglecting *radial* diffusion term. This solution (and it is not by chance that the solution has the form of impulse response of ADM) still depends upon radial coordinate but is substituted to the Eq.(1) integrated in radial direction. This averaged equation describes axial profile of mean concentration, and is solved by Fourier transform, giving short time asymptotic solution expressed again in terms of error functions. The same approach is applied also for long time asymptote giving Taylor's solution, Eq.(2).

What about turbulent flows? Analysis of tracer dispersion in fully developed turbulent flow was presented first by Taylor in 1954 using similar way of reasoning as for laminar

dispersion. Parabolic velocity profile was substituted by universal turbulent profile, and radial diffusion was based on Reynold's analogy between momentum and mass transfer. Thus the intensity of diffusion was expressed in terms of wall shear stress τ_w (or friction velocity v^*) giving dispersion coefficient in the very simple form

$$D_c = 10.1Rv^* = 10.1R\sqrt{\frac{\tau_w}{\rho}}. \qquad (4)$$

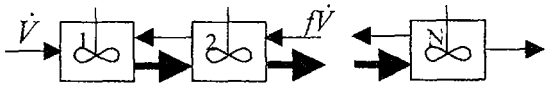
This result holds for high Reynolds numbers (Re>20000) due to the universal velocity profile which is valid only if laminar sublayer is negligibly small. Solution for the whole range of Re using experimental velocity profiles was presented by Tichacek et al (1957); he studied also the effect of wall roughness which tends to increase the axial dispersion (this is in agreement with Eq.(4)). Expression of dispersion coefficient as a function of Reynolds number can be found for example in Nauman (1983).

CFD programs have been also used for modelling turbulent flow in circular pipe. For example Ekambara and Joshi (2004) simulated stimulus response experiment in a circular pipe by Euler's method using low Reynolds number k-ε model turbulence and in-house CFD code enabling to analyse 12 different low Reynolds number models. It seems that their results predict axial dispersion coefficients which are in very close agreement with experimental data for Sc=0.27, 0.82, 1 and 1000 within the range of Reynolds number 7000 to 100000. For Re>20000 the deviation of Eq.(4) is less than 5%! This is a good message, the bad message is that in order to suppress numerical dispersion an extremely fine mesh had to be used. Problem symmetry allowed to use 2D model and results (calculated responses) are presented for meshes 100x100, ..., 100x2000. It is obvious that even with the fine mesh 100x1000 results are distorted by significant approximation error. It means that if the same problem is to be solved in 3D at least 20 millions cells have to be used (well, may be a little bit less, if a better upwind scheme is used). Thýn et al (2000) calculated turbulent, low Reynolds number flow in a pipe using different models of turbulence by Fluent and compared velocity profiles and friction factors: nearly the same (but wrong!) results were obtained by standard k-ε, RNG and RSM models, different (but also wrong) results by low Reynolds flow model, and only the simple Spalart Allmaras model was acceptable in the range Re<20000. Generally speaking the region of high Re in laminar and low Re turbulent flows is difficult to handle for CFD programs.

Several different experimental methods and tracers have been used for the ADM identification. Solution of salts (conductivity methods), dyes (colorometric methods), radiotracers and fluorescent tracers (PLIF methods) are most frequently used. Axial dispersion of heat can be also utilised, see Asbjørnsen and Amundsen (1970), who used frequency technique with pulse heating of air by a fine grid of resistive wires and recorded temperature responses by resistive thermometers arranged as a fine grid in the cross section of duct. This technique worked nice at high and also at very low Reynolds numbers flow, however underestimates dispersion coefficient within the range between Re 7000 and 30000 – this discrepancy is attributed to intermittent flows and changing velocity profile.

Parameters of ADM are identified by comparing experimentally obtained (or by CFD programs calculated) responses with analytical on numerical solution of ADM or an equivalent gradual mixing model with backflow, see Thýn et al (2000)

$$Pe_L = \frac{uL}{D_c} = \frac{2N}{1+2f}, \quad N > 6\sqrt{Pe_L} + 2 \qquad (5)$$



Comparison in the time domain is usually based upon Levenberg-Marquardt (least squares), sometimes also Nelder Mead or Rosenbrock (absolute deviations) methods. Moments and even weighted moments are preferred only as an initial estimate for optimisation routines.

3. ADM IN SELECTED UNIT OPERATIONS

Are you "in"? Standard applications of ADM for example in reactor engineering can be found in books mentioned previously, the following list is a selection of recent papers (of varying quality) concerning ADM – most of them are in fact continuations of previous research.

Dynamics of heat exchangers has been traditionally described by plug flow models, however last decade is characterized by a shift towards description of temperature profiles in heat exchanger channels by ADM. It is possible to obtain not only the axial dispersion but at the same time heat transfer coefficients (or NTU) using temperature oscillations experiments. Roetzel school is characterized by description of heat exchangers by a system of ADM equations for each pass, accompanied by equations describing heat transfer in solid walls. Laplace transform of time is applied and resulting system of ordinary differential equations is transformed to a canonical system of independent equations and solved analytically (this transformation requires numerical solution of an eigenvalue problem). Time courses of temperature responses are calculated by numerical inversion of Laplace transform. This approach has been applied either for shell and tube, Xuan&Roetzel (1993), and plate heat exchangers, Das&Roetzel (1995), Roetzel&Balzereit (1997), Luo&Roetzel (2001). Fluid temperature oscillations are modulated either by switching between cold and hot streams, Roetzel (1993) or by induction heating, Ros et al (1995), however conductivity method is also applied for independent evaluation of dispersion in HE. Temperature as a tracer is frequently used in flow-meters with pulse ohmic heating: transit time method for slow laminar flows was analyzed in terms of ADM and experimentally verified by Žitný (1993), see Thýn (2002).

Probably the highest frequency of papers is in the domain of column apparatuses. Comparison of axial dispersion coefficients (Pe ~ ReSc) in packed beds predicted by different theories with experiments is presented by doyen of packed beds, Gunn (2004). Effect of liquid phase flow direction (up and down) in hydrotreating reactor is experimentally investigated by Burkhardt (2000), influence of pressure and natural convection in upward gas flow through packed bed by Benneker (1996), and influence of Schmidt number (viscosity) by Carvalho (2003). Experiments on biological reactors with immobilised enzymes are specific by tracers (toluen, glucose) and kinetics of bioreactions, see Zarook (1998), Carrara (2003). Generally speaking most of presented correlations are quite empirical and their predictability is limited. Identification of axial and radial dispersion by CFD is usually restricted to regular packing, for example KATAPAK-S structure of catalyst particles sandwiched between corrugated sheets, van Baten et al (2001) or for static mixers, Visser et al (1999). CFD Eulerian simulations of dense and dilute phases is more frequently applied for determination of axial dispersion in bubble columns, van Baten&Krishna (2001), or bubbling fluidised bed reactors, Krishna&Baten (2001). It should be noticed that these CFD calculations make use of only slightly modified Davies-Taylor (1949) theory of bubble velocity ($u=0.71\sqrt{(2gR)}$) and that their results compare qualitatively well with Eq.(4), it is that the predicted coefficient of axial dispersion is proportional to the column diameter and liquid velocity. By the way Taylor-type analysis of axial dispersion in bubble columns was presented by Degaleesan and Dudukovič (1998) and very interesting application of two parallel ADModels for description of circulation in bubble columns is described in series of papers Degaleesan et al (1996), Gupta et al (2001) and Rados et al (2003). This is obviously a more realistic description of mixing in a bubble column, because it distinguishes core region with ascending liquid (large bubbles, low liquid holdup) and wall region with descending liquid (small bubbles, high holdup).

Pulsating flow, chaotic flows in sequence of bends, Deans vortices in coiled channels, combined effect of radial mixing and axial velocity profile in reactors with corrugated walls or baffled columns are attempts how to decrease axial dispersion especially in laminar flows.

see Howes&Mackley(1990), Castelain et al (2001), Ni et al (2002), Ye&Zhang (2002), Fitch&Ni (2003), Palma&Giudici (2003), Tu et al (2004). Axial dispersion is evaluated usually by conductivity or optical (PLIF, Particle Laser Induced Fluorescence) methods.

There exist many other technologies described by ADM (screw extruders, Puaux (2000), drum furnaces, spray towers), but surprising is the lack of applications of Taylor's dispersion for example on blood flow. Growing interest in ADM can be expected just in the field of pharmacy, bio, meso, micro and nanotechnologies, it means to applications more oriented to laminar flows. May be. Be it as it may, the next paragraph will be devoted to problems of ADM extensions in *laminar* flows.

4. ADM – EXTENSIONS IN LAMINAR FLOWS

Axial dispersion of heat is important for many operations, first of all for heat transfer apparatuses and thermal flowmeters. It is usually assumed that the axial dispersion of heat can be directly derived from the axial dispersion coefficient for mass transfer, and e.g. temperature dispersion in laminar flow can be calculated from Taylor's Eq.(2) just replacing difussivity D by temperature diffusivity a. This is not quite correct, as pointed out by Batycky et al (1993) neither in laminar nor in turbulent flows. However, their analysis is still restricted to insulated pipes and this is not the case encountered in practice. In the following we shall try to apply the Taylor's approach, this time to the heat transfer in fully developed laminar flow in circular pipe which is *not* insulated. The problem can be formulated as follows

$$\frac{\partial T}{\partial t}+2u(1-r^2)\frac{\partial T}{\partial x}=\frac{a}{r}\frac{\partial}{\partial r}(r\frac{\partial T}{\partial r}),\qquad \frac{\partial T}{\partial r}=-kT \quad \text{for } r=1 \text{ (at wall)} \tag{5}$$

In this equation T represents difference between actual temperature of fluid and a constant external temperature, the coefficient k is dimensionless heat transfer coefficient between wall and surrounding. Introducing dimensionless time τ and dimensionless axial coordinate ξ the Eq.(5) can be transformed to the new coordinate system moving with mean velocity of flow u

$$\frac{\partial T}{\partial \tau}+Pe(1-2r^2)\frac{\partial T}{\partial \xi}=\frac{1}{r}\frac{\partial}{\partial r}(r\frac{\partial T}{\partial r}),\qquad \tau=\frac{at}{R^2},\quad \xi=(x-ut)/R,\quad Pe=\frac{uR}{a} \tag{6}$$

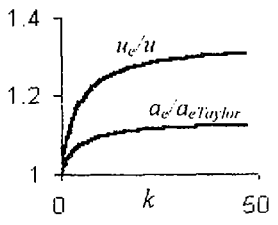
Using the same assumptions as Taylor (i.e. sufficiently long time) the solution of Eq.(6) can be approximated by

$$T=\bar{T}+\frac{Pe}{4}(-\frac{1}{3}+r^2-\frac{r^4}{2})\frac{\partial \bar{T}}{\partial \xi}+g(r)\frac{\partial^2 \bar{T}}{\partial \xi^2},\qquad \bar{T}=2\int_0^1 rTdr\,. \tag{7}$$

Mean temperature is independent of r and radial derivative of the first two terms in Eq.(7) is zero at wall (this corresponds to the case of an insulated pipe). Therefore the boundary condition (5) must be applied to the function $g(r)$, which is suggested as polynomial of 8^{th} order, and its coefficients are determined from boundary condition (5), from requirement that $\int rTdr$ is mean temperature, and first of all that after substituting temperature profile (7) into Eq.(6) the radial coordinate r disappears, i.e. the equation will have a form of ADM. This procedure is rather laborious giving ADM in the following form

$$\frac{\partial \bar{T}}{\partial \tau}+Pe\frac{12+4k}{12+3k}\frac{\partial \bar{T}}{\partial z}=-\frac{8k}{4+k}\bar{T}+\frac{Pe^2}{12}\frac{60+17k}{60(4+k)}\frac{\partial^2 \bar{T}}{\partial z^2},\qquad z=\frac{x}{R}. \tag{8}$$

This ADM reduces to the standard Taylor axial dispersion model if the tube is insulated (k=0). Increasing k increases apparent flow velocity and also the coefficient of axial dispersion only slightly as soon as k is small ($k=\alpha R/\lambda$, α is outer heat transfer coefficient, λ is thermal conductivity of liquid). Since the suggested Eq.(8) has not been thoroughly tested so far a necessity of its improvement in the future may arise.



The axial dispersion model represents a very good description of reality at turbulent flows, and also for very slow laminar flows (or for a very long times). The only principal restriction is the inequality

$$a > k\frac{\dot{V}}{L} \tag{9}$$

where the coefficient k is 0.04 according to Taylor (1954), or 0.08 according to Hunt (1977). The restriction (9) is pretty hard and in most applications with liquids, where the molecular diffusion is low, the axial dispersion calculated in laminar flows according to Eqs. (2) or (8) is too large and results are quite wrong. The situation is demonstrated in Fig.1

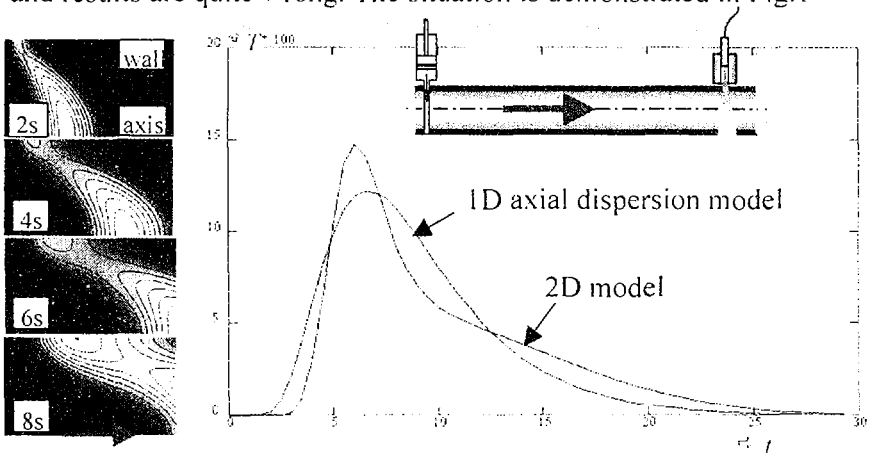


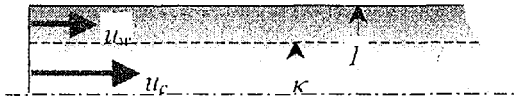
Fig.1 Response to a short impulse (mesh 60 x 40, L=1 m, $\dot{V}=\pi 10^{-5}$ m^3/s, $a=10^{-6}$ m^2/s)

Dispersion of mean calorimetric temperature ($2\int ruT\mathrm{d}r$) to a short temperature pulse at the pipe inlet (pipe is insulated) was calculated by program FEMINA, Žitný (2004), using 2D model with triangular finite elements and by 1D finite elements based upon standard model of axial dispersion with optimised dispersion coefficient. Inequality (9) is not satisfied for specified flowrate and temperature diffusivity. Fig.1 demonstrates, that the situation cannot be improved by a „better" value of the axial dispersion coefficient because the tracer dispersion in „short" times cannot be simply described by ADM. It is pity, because ADM is a very suitable model for implementation in 1D CFD programs, as compared to analytical asymptotic solutions mentioned above, which cannot be so easily generalised for modelling dispersion in complicated geometries.

Possible fall-back is to divide a pipe into several annular segments and substitute the single ADM for example by two parallel ADMs with different mean velocities u_w and u_c.

Wall region $u_w = u(1-\kappa^2)$

Core region $u_c = u(2-\kappa^2)$



Proceeding in a similar way as in deriving Eqs.(2) and (3) we arrive to the following equations describing axial dispersion of tracer in the wall region

$$\frac{\partial \bar{T}_w}{\partial \tau} + (1-\kappa^2)Pe\frac{\partial \bar{T}_w}{\partial z} = \frac{Pe^2}{48}(1-\kappa^2)^2(1+\kappa^2)\frac{\partial^2 \bar{T}_w}{\partial z^2} + \frac{4\kappa^2}{1-\kappa^2}(\bar{T}_c - \bar{T}_w) \tag{10}$$

and in the core region

$$\frac{\partial \bar{T}_c}{\partial \tau} + (2-\kappa^2)Pe\frac{\partial \bar{T}_c}{\partial z} = \frac{Pe^2}{48}\kappa^6\frac{\partial^2 \bar{T}_c}{\partial z^2} + \frac{4}{\kappa}(\bar{T}_w - \bar{T}_c), \tag{11}$$

where dimensionless time $\tau=at/R^2$, radial and axial coordinates $r=r^*/R$, resp. $z=x/R$ and also $\mathrm{Pe}=uR/a$ are the same as previously. Mean calorimetric temperature at a cross-section of pipe is calculated by means of mean temperatures in core and wall regions:

$$\bar{T} = \frac{\bar{T}_c u_c \kappa^2 + \bar{T}_w u_w (1-\kappa^2)}{u_c \kappa^2 + u_w (1-\kappa^2)} = \bar{T}_c (2-\kappa^2)\kappa^2 + \bar{T}_w (1-\kappa^2)^2 . \qquad (12)$$

It should be stressed that Eqs.(10-11) are not exact, not even asymptotically exact, because many simplifying assumptions had been done. However it is seen, that in both regions dispersion coefficients are lower than dispersion coefficient of the whole pipe, and this is what is needed, if the model should be applicable for very small diffusivities a.

The only additional parameter of the new model is κ, relative radius of core region, and can be selected in such a way, that the flowrate in both regions is the same (κ=0.541) or using criterion of the same cross-section surfaces (κ=0.707). Responses, calculated by numerical integration of Eqs.(10-11) for the same conditions as in Fig.1, are compared with the 2D solution in Fig.2

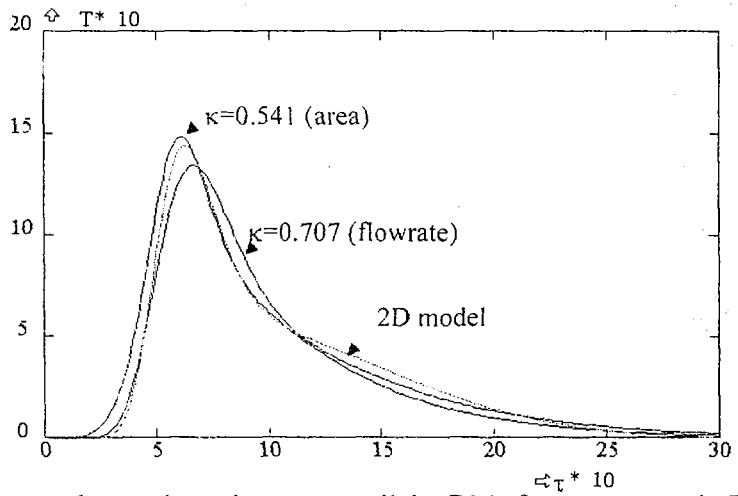


Fig.2 Responses to a short pulse using two parallel ADMs for parameters in Fig.1 (a=10^{-6} m^2/s).

It is obvious that the solutions obtained by this model are much better than standard ADM, however error increases with decreasing molecular diffusivity. For higher value of a (or lower Pe) the model of two parallel ADMs converges towards Taylor's (and exact) solution.

5. IMPLEMENTATION OF ADM IN FEM

There exist tens of programs for modeling flow, heat and mass transfer in pipelines and heat exchangers (ARROWS, FATHOM, ...). They are usually based upon control volume method and assume plug-flow in elements. ADMs have been implemented as finite elements PIPE and HEXC (heat exchanger) so far only into the program FEMINA, Žitný (2004), which together with elements CSTR (mixed tank) and PUMP allows simulation of stimulus response experiments by monitoring temperature or concentration responses in rather complicated systems. It is possible to identify model parameters by comparison with experimentally or by CFD obtained responses using integrated algorithms of collimated γ detectors and optimization routines (Levenberg-Marquardt and SOMA). FEMINA will not as general and big CFD program as FLUENT, its ambitions are rather to be an efficient RTD processor by preserving all verified features of previous generation of RTD programs developed at CTU.

CONCLUSIONS

It took 50 years from discovery of ADM to its implementation into a general purpose finite element program and the process is by far not completed. Suggested ADMs for laminar flows should be improved, e.g. their compatibility with standard heat transfer correlations.

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