

# Features of saturates mixture filtration in porous medium

V V Kachalov, I L Maikov, D A Molchanov, V M Torchinsky and  
V M Zaichenko

Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg  
2, Moscow 125412, Russia

E-mail: dmitriy.molchanov13@gmail.com

**Abstract.** Consideration is given to the filtration process of the two-phase multicomponent mixture in the porous. It is shown that “mixture-porous medium” system becomes self-oscillating one during filtration process under special conditions when there is a region of retrograde condensation on the phase diagram of the mixture. A mathematical model of the hydrocarbon mixtures filtration process of the methane series has been developed and a computer program for calculating hydrodynamic and thermodynamic characteristics of this process under isothermal conditions with phase transitions has been created. Consideration is given to the basic mechanisms influencing the filtration dynamics. Limits of the model applicability are discussed. Condition range for occurring self-oscillatory properties in “mixture-porous medium” system is determined by medium permeability, viscosity of the mixture, initial and boundary filtration conditions. Experimental filtration research of mixtures “methane-n-butane”, “methane-propane-butane”, “methane-pentane” under the thermodynamic conditions corresponding retrograde condensation region on the phase diagram have shown validity of this model. It is argued that any multicomponent mixture having a retrograde condensation region on the phase diagram appears as self-oscillating system under right conditions.

## 1. Introduction

Self-oscillating systems are widely studied both in physical and mathematical aspects. These studies, in particular, have shown that processes in any self-oscillating system are described by nonlinear differential equations, which complicates theory of these processes significantly [1, 2].

Transients in such systems approach the steady state flow regime over time in different ways depending on the system properties and forces acting. Self-oscillating systems are able to generate sustained oscillations. Nature of self-oscillations can be different: mechanical, thermal, electromagnetic and chemical ones.

Various mathematical apparatus are required for an accurate quantitative description of different self-oscillating systems. Nevertheless, one can imagine a scheme, which is common to all self-oscillatory systems and qualitatively describes properties of the system. Self-oscillations require source of permanent (non-periodic) impact and nonlinear controller, which converts direct impact to varying one, that “pumps” the oscillating system element, which control the regulator operation by feedback setting the phase and the frequency of its actions.

It is shown than the system “two-phase fluid-porous medium” is able to relegate to the class of self-oscillating systems in some circumstances. Steady periodic flow regime, which is one of



the properties of self-oscillating systems, could occur when there is a retrograde condensation region on the phase diagram and phase permeabilities are significantly different.

Pressure gradient is an energy source causing filtration in this case. The process of retrograde evaporation is a nonlinear system element, and significant difference in the permeability values of the vapor and liquid phases serves as the feedback.

## 2. Filtration regimes of hydrocarbon mixtures

A mathematical model of the filtration process of the methane series hydrocarbon mixtures, whose phase diagram had been more fully studied [3], has been developed and a computer program for calculating hydrodynamic and thermodynamic characteristics of this process under isothermal conditions with phase transitions has been created. Conditions, under which “mixture–porous medium” system shows self-oscillating properties, have been found. The basic mechanisms influencing filtration process dynamics are a significant difference in the permeability values and the phenomenon of liquid phase retrograde evaporation in the retrograde region on the phase diagram with pressure increase. There are different flow regimes of mixture filtration depending on boundary conditions:

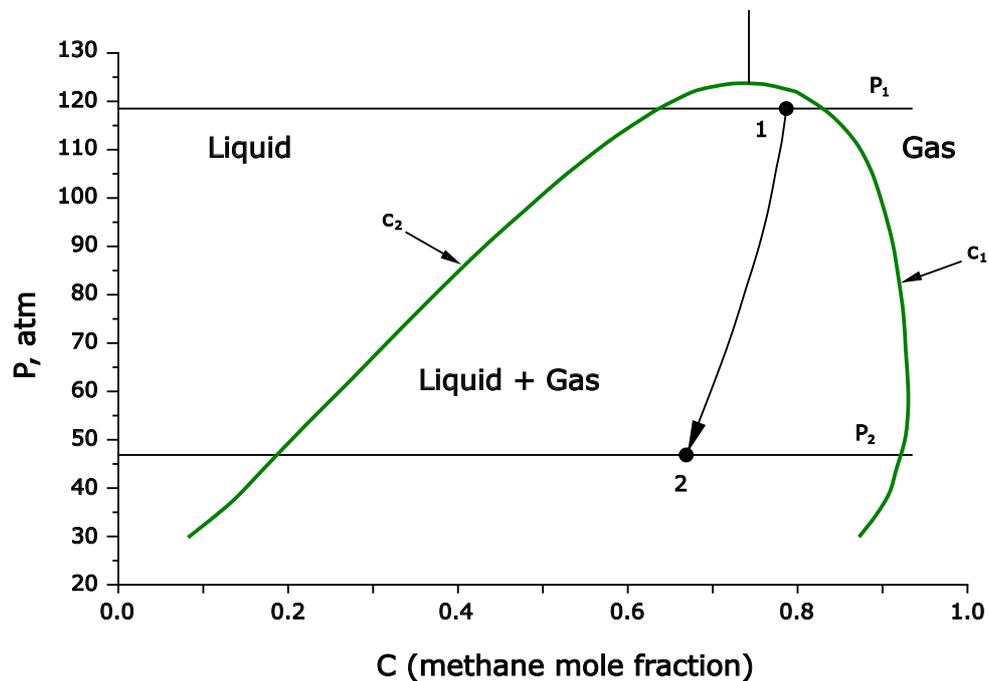
- Two-phase filtration with a constant flow rate of liquid and gas phases, wherein pressure gradient is in the region below the pressure of maximum condensation;
- Two-phase filtration with cyclic flow rate change of liquid and gas phases, when maximum pressure of the pressure gradient is in the region between supercritical pressure and pressure of maximum condensation (self-oscillating regime);
- Fluid filtration in the supercritical state with constant flow rate, pressure gradient is in the region above pressure of maximum condensation.

A computer program has been developed for the experimental verification of the mathematical model and corresponding parameters of the experimental physical model has been defined by numerical simulation. Experiments of the hydrocarbon mixtures filtration of the methane series (“methane–n-butane”, “methane–propane–butane”, “methane–pentane”) in the thermodynamic conditions corresponding with retrograde region on the phase diagram has been carried out [4]. Dynamics of flow rate and pressure gradient and composition of the liquid and gas phases have been studied. Experiment have been carried out both with constant temperature and with changing temperature conditions during filtration process. The results of physical simulation have confirmed the conclusions which have followed mathematical model – thermodynamic filtration conditions and filtration properties of the porous medium, in which “mixture–porous medium” system shows self-oscillation properties, have been found experimentally.

Developed theory of the phenomenon and results of the physical simulation have allowed us to conclude that any multicomponent mixture having a region of retrograde condensation on its phase diagram is self-oscillating system in appropriate conditions.

## 3. Mathematical model

Filtration process of the methane series hydrocarbon mixture in the porous medium results in issues with two-phase flow with phase transitions distributed in the space, which occur not on a separate surface (such as classical Stefan problem) but in spatial domain. The phase diagram of such a mixture contains a retrograde region, in which retrograde liquid may form while pressure drops to the pressure of maximum condensation, and it will evaporate with further pressure decrease [5]. Besides unusual phase diagram, notice should be taken of additional features in the behavior of phase permeability functions occurring during hydrocarbon mixture filtration in porous medium. Values of relative permeabilities for gas and liquid phases can be close to zero. Thus, it is necessary to identify new physical laws which are specific for hydrocarbon mixture



**Figure 1.** The phase diagram of “methane–n-butane” binary mixture at a temperature of 310 K,  $c_1(P)$  – curve of equilibrium concentration in the gas,  $c_2(P)$  – curve of equilibrium concentration in the liquid.

filtration process with particular mixture composition, thermal fluid properties, properties of porous medium and occurring phase transitions.

Consideration is given to one-dimensional two-phase non-stationary filtration of two-component system under isothermal condition with assumption that pressures in the phases are equal provided by phase equilibrium (the characteristic times of phase transitions are much smaller than hydrodynamic ones). The phase diagram of “methane–n-butane” mixture is shown on figure 1.

State 1 on the phase diagram corresponds to the input boundary condition  $P_1$ , state 2 – output boundary condition  $P_2$ . The hydrocarbon mixture passes from state 1 to state 2 by filtration. We denote the distance, at which the pressure changes from  $P_1$  to  $P_2$ , as  $L$ . Filtration process in one-dimensional case is described by the equations of mass balance for each component written in divergence form with respect to the molar density, components are chemically neutral:

$$m \frac{\partial}{\partial t} \left( \sum_j c_{ij} n_j s_j \right) + \frac{\partial}{\partial x} \left( \sum_j c_{ij} n_j U_j \right) = 0, \quad (1)$$

where  $m$  – the porosity; index  $i$  corresponds to the component ( $\text{CH}_4$ ,  $\text{C}_4\text{H}_{10}$ ); index  $j$  correspond to the phase (1 – gas, 2 – liquid),  $c_{ij}$  – mole fraction of the  $i$ -th component in the  $j$ -th phase of the two-phase mixture;  $n_j$  – molar density of  $j$ -th phase;  $s_j$  – volume fraction of the  $j$ -th phase in the mixture;  $U_j$  – speed of  $j$ -th phase;  $t$  – time;  $x$  – coordinate position.

Momentum conservation equations are written in the approximation of Darcy’s law:

$$U_j = -k \frac{f_j(s_j)}{\mu_j} \frac{\partial P}{\partial x}, \quad (2)$$

where  $k$  – absolute permeability;  $\mu_j$  – dynamic viscosity of the  $j$ -th phase;  $P$  – pressure;  $f_j$  – relative permeability of the  $j$ -th phase.

Equations of state are written for gas and liquid phases:

$$P = n_j z_j RT, \quad (3)$$

where  $z_j$  – compressibility factor of the  $j$ -th phase;  $R$  – gas constant;  $T$  – mixture temperature.

The equilibrium concentrations of the components in both phases depend on the pressure and satisfy the following condition:

$$\sum_i c_{ij}(P) = 1, \quad (4)$$

Volume fractions of the phases are related by following condition:

$$\sum_j s_j = 1. \quad (5)$$

The equation system is supplemented by boundary and initial conditions. Functional dependencies  $f_1(s)$  and  $f_2(s)$  are the following [6]:

$$f_1(s) = \begin{cases} 0, & \text{if } 0 \leq s \leq s_{1k}; \\ [(s - s_{1k})/(1 - s_{1k})]^{3.5}, & \text{if } s_{1k} \leq s \leq 1; \end{cases} \quad (6)$$

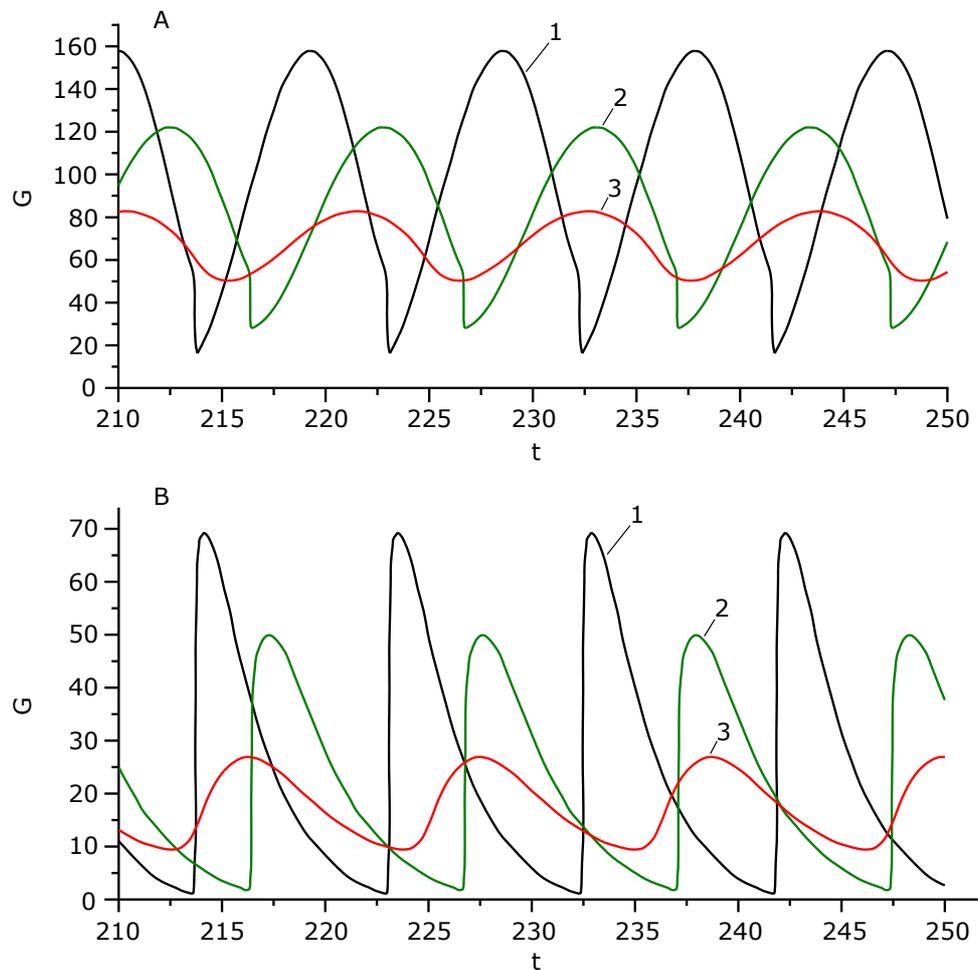
$$f_2(s) = \begin{cases} [(s_{2k} - s)/(s_{2k})]^{3.5}, & \text{if } 0 \leq s \leq s_{2k}; \\ 0, & \text{if } s_{2k} \leq s \leq 1, \end{cases} \quad (7)$$

where  $s_{1k}$  and  $s_{2k}$  – parameters. In the paper, this parameters have the following values:  $s_{1k} = 0.35$  and  $s_{2k} = 0.8$ . There are regions in which each of these functions becomes equal to 0, i.e. its velocity is 0. It is the main feature of these dependencies. Physically it means that there are some areas where the phase is not continuous, but it is present in the form of bubbles and droplets which are stationary.

#### 4. Results of the mathematical simulation

Figures 2 and 3 show calculation results of hydrocarbon mixture flow rate and pressure changes over time for one of the filtration regimes. Inlet pressure  $P_1$  is steady and equal to 120 atm.

The filtration process is a sequence of stationary processes (by pressure), differing by position of the pressure front, at which gas saturation reaches  $s_{1k}$ . Consideration of possible filtration regimes is given. Flow in the system is due to given pressure gradient, specific pressure values determine part of phase diagram and all other properties of oscillating system. Flow rate and upstream conductivity decrease due to intrinsic properties of the system (condensate dropout in retrograde region, reducing of the gas permeability). Also reducing of pressure gradient can decrease flow rate. Flow of gas phase stops. Although liquid density is above gas one, but liquid velocity is significantly lower than gas one, and therefore system energy decreases (stage of returning of energy to external source) and reaches a minimum value, wherein  $\Delta E_{\text{out}}^{\text{max}}$  is the maximum amount of given energy in the period. Then flow rate begins to increase (condensate evaporation, increase of gas permeability and upstream conductivity). Increase of system energy begins as a result of the involvement of stopped gas phase into the movement (stage of energy extraction from an external source) and system energy reaches a maximum value, wherein  $\Delta E_{\text{in}}^{\text{max}}$  is the maximum amount of added energy from in the period. If  $\Delta E_{\text{out}}^{\text{max}} - \Delta E_{\text{in}}^{\text{max}} > 0$ , then energy is taken from the system during each oscillation period and damped oscillations take place. If on the contrary,  $\Delta E_{\text{out}}^{\text{max}} - \Delta E_{\text{in}}^{\text{max}} < 0$ , then system energy and oscillations increase. Thus, the system is self-oscillating one [7]. So, there is a power supply of the system, moreover



**Figure 2.** Time-dependences of gas (A) and liquid (B) flow rate (in relative units) for different outlet pressures  $P_2$ : 1 – 72.5 atm, 2 – 77.5 atm, 3 – 82.5 atm.

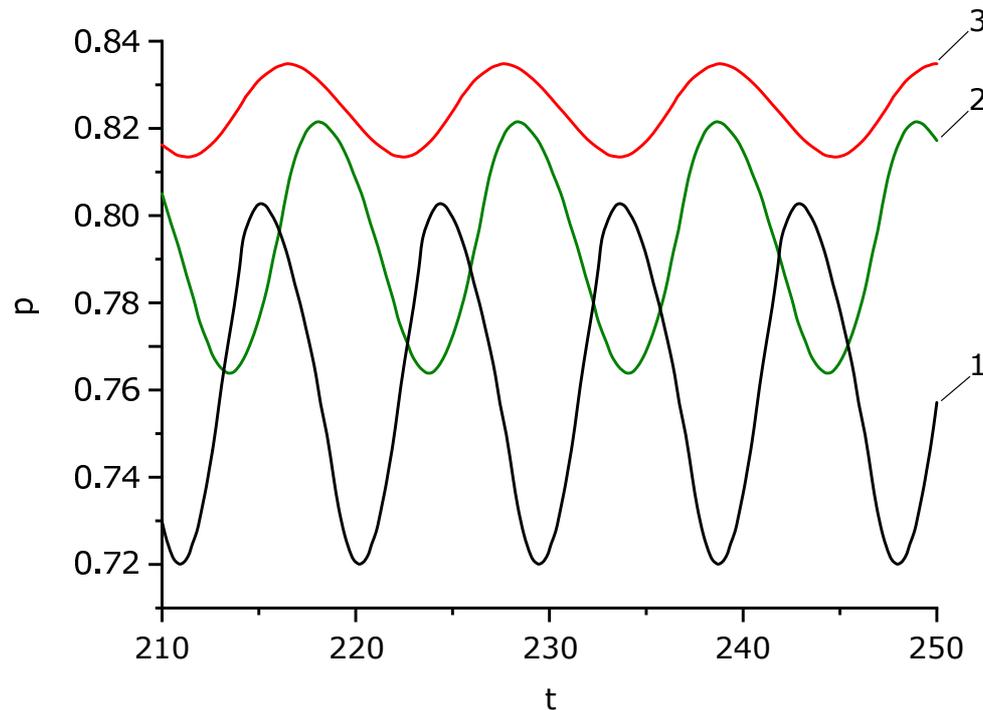
the supply of energy does not occur at random but it is carried out by controlling mechanism actuated by the system. Control mechanism operates as a feedback between oscillating system and energy source and provides energy input in the required time of the oscillation period. A similar mechanism will occur during the filtration of any fluids, whose phase diagram contains a region of retrograde condensation, under appropriate conditions.

## 5. Conclusions

Consideration is given to the filtration process of hydrocarbon mixtures with the features on the phase diagram in porous medium. The mathematical model characteristics of this process under isothermal conditions with phase transitions has been developed and computer program for calculating of hydrodynamic and thermodynamic characteristics has been created. Experimental studies confirm numerical simulation in essence [8]. The conditions, under which “mixture–porous medium” system shows self-oscillating properties, have been found.

## Acknowledgments

This work was supported by the Russian Science Foundation (project No. 14-50-00124).



**Figure 3.** Time-dependences of pressure (in relative units)  $p = P/P_1$  ( $x = 0.5$ ) for different outlet pressures  $P_2$ : 1 – 72.5 atm, 2 – 77.5 atm, 3 – 82.5 atm.

### References

- [1] Blacker O 1969 *Analysis of Nonlinear Systems* (Moscow: Mir)
- [2] Migulin V 1978 *Basic Theory of Oscillations* (Moscow: Mir)
- [3] Batalin O, Brusilovskiy A and Zaharov M 1992 *Phase Equilibria in Systems of Natural Hydrocarbons* (Moscow: Nedra)
- [4] Zaichenko V M, Maikov I L and Torchinskii V M 2013 *High Temperature* **51** 776–784
- [5] Zaichenko V M, Maikov I L, Torchinskii V M and Shpilrain E E 2009 *High Temperature* **47** 701–706
- [6] Vafina N, Zakirov S and Jufin P 1988 *Proceedings of the Universities, Oil and Gas* **9** 56–63
- [7] Magnus K 1982 *Oscillations* (Moscow: Mir)
- [8] Zaichenko V M, Maikov I L, Smolkin A, Sokol G and Torchinskii V M 2012 *Instruments and Experimental Techniques* **5** 95–98