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# Water purification of organic inclusions in a reverse flow filtration combustion reactor

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#### ABSTRACT

A new technology of water purification of organic inclusions or solutes by using a filtration combustion reactor with alternating flow direction is investigated numerically. An acetone aqueous solution served as a model liquid. The lean combustibility limit (LCL) was determined for a wide range of flow rates, different values of the heat exchange coefficient, length of the reactor, and for different sizes of packed bed particles. It is demonstrated that thermal insulation has the strongest effect on the LCL. A minimum concentration of acetone in water that provides self-sustaining combustion may be as low as  $c_{\min} = 13.5\%$  for the reactor length L = 0.4 m. The nonmonotonic behavior of the LCL dependence of the flow rate for short reactors and coarse particles has been demonstrated. The physical aspects of this feature was discussed.

### 1. Introduction

Water purification of organic inclusions is of current interest for industry. The problem becomes more urgent because of the aggravation of ecological problems and reduction in the reserves of high quality pure water in the world.

There are numerous methods of water purification of organic components. These methods include sorption, distillation, thermal, biochemical, and electrochemical decomposition of emulsions, and others [1–3]. All of them are comparatively expensive and in some cases generate secondary pollutions which require utilization.

Recently it has been proposed [4,5] to use the method of filtration combustion for oxidative purification of water. The principles of filtration combustion, also called "excess enthalpy combustion" are presented in [6]. In a filtration combustion system one can locally obtain a temperature that exceeds the adiabatic temperature of the combustion of a mixture. The methods of filtration combustion are applied to the combustion of extra-lean and extra-rich gas fuel mixtures [7].

Recently oxidative clearing of water from acetone in a "tube-intube" type reactor was demonstrated experimentally [5] and investigated numerically [8]. A minimal concentration of acetone in water, which provides self-sustainable combustion (lean combustibility limit), was determined for the typical parameters of a laboratory scale reactor. The value of the lean combustibility limit

\* Corresponding author. E-mail address: ivkozn@hmti.ac.by (I.A. Koznacheev). is most sensible to the coefficient of heat losses and comprises a value of 14% (mass) for typical parameters of such a reactor.

Another type of reactor that realizes the excess enthalpy regime is a reactor with an alternating flow direction, also called a regenerative type reactor. The main advantage of the reactor of this type is the excellent scale up capability, associated with its 1D geometry. Therefore, this type of reactor deserves a profound study.

In this paper, we perform systematic computer simulation of the process of acetone–water mixture oxidative purification in a reverse flow reactor. As to its methods and aim the paper is quite similar to [8]. The main concern of the present investigation is the determination of a minimal concentration of acetone that provides self-sustained thermal oxidation.

### 2. Statement of the problem

Let us consider a regenerative reactor with a reversible flow, Fig. 1. The reactor is filled with a packed bed of ceramic balls and is thermally insulated from outside. Water contaminated with organic compounds is fed, along with air, alternatively from the ends of the reactor. The water and organic components evaporate in the hot zone and burn up. The combustion products heat the porous medium and leave the system. Periodical switching of the flow direction is provided by a pair of two-way valves. The flow direction is switched after the combustion front travels over the center of the reactor.

A generalized volume-averaged model [9,10] was used to simulate the system. The set of equations included: continuity and pressure equations for the gaseous phase, conservation

Nomenclature					
С	concentration of acetone in liquid	$T_{\rm ext}$	temperature of ambience (K)		
$c_p$	specific heat capacity of gas mixture (J/(kg K))	u	filtration velocity vector (m/s)		
$c_{w}$	mass concentration of liquid (kg/m³)	$Y_i$	mass fraction of the ith gas component		
$c_{wat}$	mass fraction of water in working mixture	Z	longitudinal coordinate (m)		
$c_{\min}$	minimal concentration of organic component for auto-				
	thermal process	Greek	Greek symbols		
D	gas diffusivity tensor	$\alpha_{\rm vol}$	volumetric interphase heat transfer coefficient		
d	reactor tube diameter (m)		$(W/K/m^3)$		
$d_0$	packed bed particle diameter (m)	α	coefficient of heat exchange with the ambience		
g	specific mass flow rate (kg/(m <sup>2</sup> s))		$(W/K/m^2)$		
$h_i$	mass enthalpy of ith chemical component (J/kg)	λ	solid conductivity (W/m/K)		
$I_{ev}$	evaporation rate $(kg/(m^3 s))$	$\mu$	gas viscosity (Pa s)		
$k_0, k_1$	filtration permeabilities	Λ	heat conductivity tensor		
L	reactor length (m)	ho	density (kg/m³)		
M	average molecular weight of gas (kg/mol)	$\dot{ ho}_i$	mass generation of ith component due to chemical reac-		
$M_{w}$	molar mass of liquid (kg/mol)		tions		
m	porosity				
p	pressure (Pa)	Subscr	Subscripts		
$p_{sat}$	saturated vapor pressure (Pa)	ad	adiabatic		
Q	heat (J)	g	gas		
r	radial coordinate (m)	i	ith component of a gas		
R	molar gas constant (J/(mol K))	S	solid		
T	temperature (K)	liq	iquid		
$T_{0}$	temperature of the working mixture at inlet (K)				

equations for the gaseous components, energy conservation equations for the gas and solid, and the ideal gas state equation:

$$\frac{\partial \rho_{\mathbf{g}}}{\partial t} + \nabla(\rho_{\mathbf{g}} \mathbf{u}) = I_{\mathbf{ev}},\tag{1}$$

$$-\nabla p = \frac{\mu}{k_0} \mathbf{u} + \frac{\rho_{\rm g}}{k_1} |\mathbf{u}| \mathbf{u}, \tag{2}$$

$$\rho_{\rm g} \frac{\partial Y_i}{\partial t} + \rho_{\rm g} \mathbf{u} \nabla Y_i - \nabla \rho_{\rm g} \mathbf{D} \nabla Y_i = \dot{\rho}_i, \tag{3}$$

$$\rho_{\rm g} c_p \frac{\partial T_{\rm g}}{\partial t} + c_p \rho_{\rm g} \mathbf{u} \nabla T_{\rm g} - \nabla (\mathbf{\Lambda} \nabla T_{\rm g}) = \frac{\alpha_{\rm vol}}{m} (T_{\rm s} - T_{\rm g}) - \sum_i h_i \dot{\rho}_i, \qquad (4)$$

$$(1-m)\rho_{s}c_{s}\frac{\partial T_{s}}{\partial t}-\nabla(\lambda\nabla T_{s})=\alpha_{vol}(T_{g}-T_{s}), \tag{5}$$

$$\rho_{\rm g} = \frac{pM}{RT_{\rm g}}.\tag{6}$$

The governing equations were supplemented with the conservation equation for the mass of the liquid injected into the system:

$$\frac{\partial c_{\rm w}}{\partial t} + u_{\rm w} \frac{\partial c_{\rm w}}{\partial z} = -I_{\rm ev},\tag{7}$$

where  $u_{\rm w}$  is the constant mean velocity of the liquid, equal to the ratio of the specific flow rate to the liquid concentration in the solid phase. Eq. (7) is written twice: for water and acetone separately.

The evaporation rate per volume unit was evaluated as follows:

$$I_{\text{ev}} = \frac{3(1-m)c_{\text{w}}}{d_{0}c_{\text{w.0}}\sqrt{2\pi\frac{RT_{\text{s}}}{M_{\text{w}}}}}(p_{\text{sat}} - p). \tag{8}$$

The saturated vapor pressure was calculated by a semi-empirical formula [8].

The gas flow rate and composition at the inlet were fixed. A constant pressure at the outlet was assigned. The Newtonian heat exchange with the ambience was used.

$$-\lambda \frac{\partial T_{s}}{\partial r}\Big|_{r=0.5d} = \alpha (T_{s}|_{r=0.5d} - T_{\text{ext}}). \tag{9}$$

Another condition used is the adiabatic one.

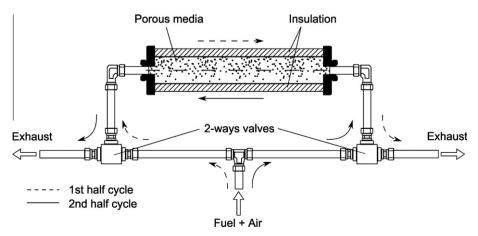


Fig. 1. Diagram of the reverse flow filtration combustion reactor.

Heat transfer and diffusion coefficients are expressed analytically [8].

A simple one-reaction chemical mechanism of acetone combustion was used in the calculations:

$$CH_3COCH_3 + 4O_2 \Rightarrow 3CO_2 + 3H_2O$$

It was suggested that the reaction is of the first order for acetone and oxygen. The numerical values of the activation energy and of the pre-exponential factor were taken from [11].

The 2DBurner software package [12] was used for numerical solution of the problem. Most of calculations were taken for onedimensional problem statement. Model and software package verification was performed in [8].

#### 3. Results and discussion

The simulation has reproduced the unsteady process of reactor operation, a periodical change of the flow direction and the establishment of a cyclic regime. The combustion process was initiated by preheating a short zone of the porous medium up to  $T=1500~\rm K$ . The cyclic thermal and temporal parameters were settled after approximately 5–9 cycles of flow switching. All the parameters discussed in the paper relate to the settled cyclic operation regime of the reactor. A maximum temperature of the porous medium corresponds to the moment when the combustion front passes over the middle of the reactor.

The values of the parameters corresponding to the standard simulation case (standard system) are presented in Table 1.

The character of the establishment of the cycle for different concentrations of organics in water is shown in Fig. 2.

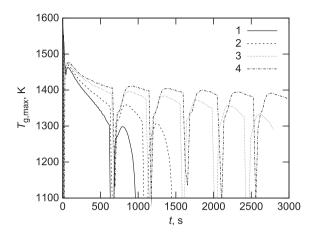
The graphs demonstrate that the combustion wave can be extinguished during a relatively large number of periods of reactor operation. This is due to the fact that the extinguishment of the filtration combustion wave near the concentration limit occurs slowly at a distance much greater than the reactor length.

The specific feature of the system considered is the presence of an evaporation wave traveling within the porous medium after switching of flow direction. The dynamics of temperature profiles after switching is shown in Fig. 3. The propagation of the evaporation wave which lowers the temperature is clearly demonstrated by curves 2 and 3. When the evaporation wave reaches the main front of the filtration combustion wave, the latter becomes steeper.

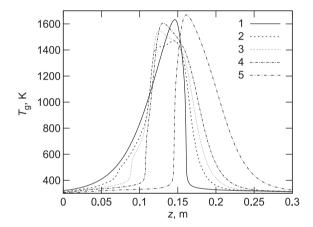
The maximum temperature in the combustion wave is one of the most important characteristics of filtration combustion system. The dimensionless ratio of a maximum temperature to adiabatic combustion temperature  $(T_{\rm max}-T_0)/(T_{\rm ad}-T_0)$  characterizes heat

**Table 1** Standard system parameters.

Description	Symbol	Value
Specific heat capacity of the packed bed material	c <sub>s</sub>	1000 J/(kg K)
Packed bed material density	$ ho_{s}$	1250 kg/m <sup>3</sup>
Porosity	m	0.4
Reactor size		
Length	L	0.4 m
Tube diameter	d	0.04 m
Liquid density		1000 kg/m <sup>3</sup>
Size of solid particles	$d_0$	5 mm
Specific flow rate of liquid	$g_{\rm liq}$	$0.1 \text{ kg/(m}^2 \text{ s})$
Mean liquid velocity	$u_{\rm w}$	1.4 mm/s
Concentration of liquid in the solid phase at the inlet	<i>c</i> <sub>w.0</sub>	71 kg/m <sup>3</sup>
Concentration of acetone in liquid	С	0.4
Temperature of incoming air and of the ambience	$T_0$ , $T_{\text{ext}}$	300 K
Coefficient of heat exchange with the ambience	α	$4 \text{ W}/(\text{m}^2 \text{ K})$
Specific flow rate of air		Stoichiometric
Pressure at the outlet		1 atm



**Fig. 2.** Settlement of the temperature cycle in the system. Concentration of the organic components in water, c: 1 - 14.0%, 2 - 14.4%, 3 - 14.8%, 4 - 15.0%.



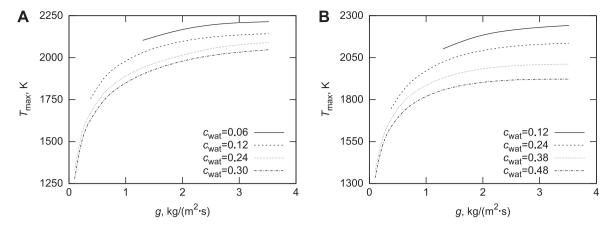
**Fig. 3.** The dynamics of temperature field after switching of flow direction. Time from the moment of switching: 1 - 0 s, 2 - 10.45 s, 3 - 16.15 s, 4 - 23.15 s, 5 - 48.05 s

recuperation in the wave. Calculations show that the dependence of a maximum temperature in the front on the flow rate (Fig. 4A) is a monotonous function similar to the case of filtration combustion of gases [13]. The maximum temperature increases with the heat content of the mixture (concentration of organics). The rate of growth of  $T_{\rm max}$  is a permanently decreasing function. This behavior is attributed to a pressure drop in the system, chemical kinetics and other factors generally discussed in [13].

Let us investigate the features of the system determined by the presence of liquid phase at the reactor inlet. Consider two cases. A two-phase water–acetone–air mixture (acetone–air ratio corresponds to chemical stoichiometry) is fed to the reactor in the first case at  $T_0$  = 300 K. A one-phase gaseous fuel mixture of the same chemical composition is fed at  $T_0$  = 375 K in the second case. In the latter case the enthalpy of the mixture is higher due to the elevated temperature and phase transition energy. The dependence of the maximum temperature in the front on the flow rate at different water contents in the working mixture  $c_{\rm wat}$  was calculated for both cases, Fig. 4.

It is easy to see that the maximum temperature is higher in the second case for the same water content, Fig. 4. This is explained by the higher enthalpy of the mixture.

The upper curves (solid lines) in Fig. 4 are limited from the side of lower flow rates. This corresponds to the end of the regime of downstream propagation of the combustion wave, but stabilization at the reactor inlet. The character of the  $T_{\text{max}} = f(g)$  functional



**Fig. 4.** The dependence of the maximum temperature on the specific mass flow rate of the working mixture. (A) The two-phase mixture at the inlet,  $T_0 = 300 \text{ K}$ ; (B) The one-phase (gaseous) mixture at the inlet,  $T_0 = 375 \text{ K}$ .

dependence remains the same for both cases. However, the value of  $C_{\rm rec} = \frac{T_{\rm max} - T_0}{T_{\rm ad} - T_0}$  that characterizes heat recuperation efficiency in the system is quite different, Fig. 5. The curves are built for identical working mixture compositions (stoichiometry acetone-air, mass fraction of water – 30%). In the case of the two-phase mixture feeding, this value is up to two times higher compared to the case of the one-phase feed. This is mainly connected with the change in the front structure due to the phase transition steeping front. Generally the increase in the heat recuperation efficiency makes it possible to achieve higher temperature in the system (the input enthalpy being constant) and obtain a lower concentration combustion limit (or lean combustibility limit). From this viewpoint, organization of two-phase feeding may ensure better combustion of extra-lean fuels.

The lean combustibility limit (LCL) is an important characteristic of the system, as far as it defines the scope of the use of the technology and the class of compounds that can be oxidized in the auto-thermal process.

To determine the LCL, we simulated a corresponding experimental cycle. At fixed values of the reactor parameters, combustion was initiated and calculations were carried out until a periodic regime of operation was established. Then, keeping all the parameters constant, we reduced the content of acetone in the mixture, with the stoichiometric ratio of air to acetone being conserved. Calculation was continued until a new periodic regime of operation was established. A step-by-step reduction of the concentration of acetone was carried out until combustion quenching. The arithme-

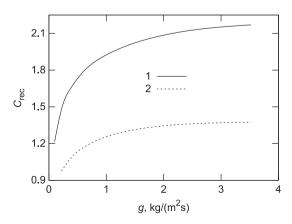
tic mean of two latter concentrations is taken as the LCL for the given mode of reactor operation.

The LCL was determined as a function of the specific liquid flow rate, of the heat exchange coefficient  $\alpha$  (associated with the thickness of insulation), of the length of the system, and of the size of packed bed particles.

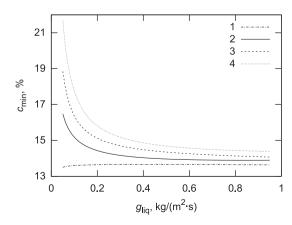
According to the general concepts of filtration combustion [9,14], the main factors that determine the combustibility limit in such a system are the heat loss to the environment and the recovery of heat by means of the gas–solid heat exchange. For the considered reactor the combustibility limit can also be affected by the transition regime after the switching of flow direction (Fig. 3). The dependence of the LCL on the specific flow rate of liquid at different values of the heat exchange coefficient is shown in Fig. 6. The curves calculated for nonadiabatic cases approach the adiabatic one at higher  $g_{liq}$  as far as the growth of the flow rate is equivalent to the increase in the energy input to the system  $Q_{inp}$  and decrease in the dimensionless side loss  $Q_{loss}/Q_{inp}$  to input energy ratio.

The dependence of the LCL on the specific flow rate for reactors of different lengths (diameter of the reactor is constant) was obtained numerically, Fig. 7.

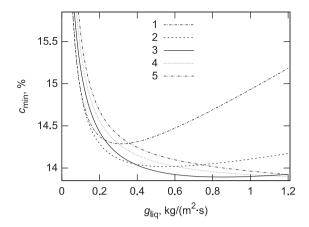
Fig. 7 demonstrates a nonmonotonic character of the LCL dependence on  $g_{liq}$ . Note that such a dependence of the LCL on flow rate takes place also for systems of filtration combustion of gases (FCG) without phase transition [15]. However, for systems without phase transitions the intersection of the curves  $c_{min} = f(g_{liq})$ , built



**Fig. 5.** Dependence of  $C_{\text{rec}}$  on the flow rate for the two-phase (1) and one-phase (2) fuel feed.  $T_0$  = 300 K for the first case,  $T_0$  = 375 K for the second case.



**Fig. 6.** The dependence of the concentration limit on the mass flow rate of liquid for different heat exchange coefficients:  $1 - \alpha = 0$  (adiabatic case),  $2 - \alpha = 5.113$ ,  $3 - \alpha = 10.226$ ,  $4 - \alpha = 15.339$  W/(m<sup>2</sup> K). Other parameters have standard values.



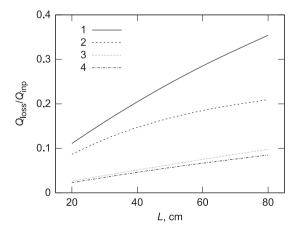
**Fig. 7.** The dependence of the concentration limit on the mass flow rate of liquid for different reactor lengths: 1 - L = 20 cm, 2 - L = 30 cm, 3 - L = 40 cm, 4 - L = 60 cm, 5 - L = 80 cm. Other parameters have standard values.

for different reactor lengths, was not observed. The intersection is connected with the inversion of LCL dependence on the reactor length: at high flow rates the LCL is lower for longer reactors (which is consistent with the known regularities for FCG, [15]); at low flow rates the LCL is lower for shorter reactors, which is a new regularity peculiar to systems with phase transition.

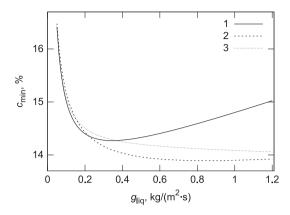
This regularity is of complex physical nature. At low flow rates heat losses at side walls  $Q_{\rm loss}$  substantially influence the LCL. In its turn the  $Q_{\rm loss}$  value is connected with the width of the hot area of the combustion wave. This width is smaller for the system with phase transition. Reduction of the hot area of the system with phase transition is of twofold nature: (1) sharpening of the front due to its «binding» to the phase transition temperature, (Fig. 3), (2) speedup of the downstream movement of the front, and a corresponding decrease in the flow switching period.

The dependence of the heat losses through the walls normalized to the heat content of the incoming mixture as a function of the reactor length is given in Fig. 8. This dependence adds to the understanding of the nature of the inversion.

For both cases of low and high flow rates the  $Q_{loss}$  values are lower for the case of two-phase mixture combustion (curves 2, 4). For both cases  $Q_{loss}$  grows with increase in the reactor length. However, for high flow rates the ratio  $Q_{loss}/Q_{inp}$  is much smaller. Therefore heat losses with the exhaust gas are of major importance. These losses are greater for the shortest reactors. Inversion



**Fig. 8.** Normalized heat losses through the walls as a function of the reactor length. 1,  $3 - T_0 = 375 \text{ K}$ , 2,  $4 - T_0 = 300 \text{ K}$ . 1,  $2 - g = 0.3 \text{ kg/(m}^2 \text{ s)}$ , 3,  $4 - g = 1.5 \text{ kg/(m}^2 \text{ s)}$ , c = 0.2, other parameters have standard values.



**Fig. 9.** The dependence of the concentration limit on the mass flow rate of liquid for different particle sizes:  $1 - d_0 = 1$  cm,  $2 - d_0 = 0.5$  cm,  $3 - d_0 = 0.25$  cm. Other parameters have standard values.

takes place because of transition from the regime, where the LCL is determined by heat losses through the walls, to the regime, where the LCL is determined by heat losses with the exhaust gas.

The dependence of the lean combustibility limit on the specific liquid flow rate for different values of the size of particles,  $c_{\min} = f(g_{\text{liq}}, d_0)$ , is shown in Fig. 9. The function drops at lower flow rates following the general regularity of the reduction of the dimensionless side losses  $Q_{\text{loss}}/Q_{\text{inp}}$ . Since the heat losses through the walls are almost independent of particle size, the LCL does not depend on the particle size at low flow rates. At higher flow rates the factor of reduction of dimensionless heat recirculation  $Q_{\text{rec}}/Q_{\text{inp}}$ , starts to play a dominating role especially for large sizes of particles. The concurrency of both factors determine the nonmonotonic behavior of the function  $c_{\min} = f(g_{\text{liq}})$ . The same regularity takes place for the case of gas filtration combustion [15].

#### 4. Conclusions

The new technology of water purification of organic inclusions or solutes by means of filtration combustion has been substantiated and investigated numerically. The essence of the technology is in simultaneous evaporation of a liquid and combustion of organic components within an inert porous medium in a regenerative reactor. Acetone solution in water was used as a model liquid.

The standard model of filtration combustion based on heat and mass conservation equations for a porous medium and the gas phase, taking into consideration chemical reactions in the gas phase, was used. The standard model was supplemented with the equation of mass conservation for the liquid phase and phase transition. The model has been verified using the experimental data of thermometric measurements of the system [8]. One-dimensional simulation of the system has been carried out.

The effect of the presence of a liquid phase on the thermal characteristics of the reverse flow reactor has been studied qualitatively. It was found that in the case of feeding of a two-phase mixture, one can obtain higher temperatures in the wave compared to feeding gas of the same total enthalpy. The physical reason for this is the restructuring of the front in the presence of a phase transition, acceleration of the downstream movement, and the increase in the heat recirculation factor.

The dependence of the lean combustibility limit on the specific liquid flow rate at small lengths of the reactor and large sizes of particles are nonmonotonic (see Figs. 7 and 8). This is due to the fact that, at low flow rates the major influencing concentration limit factor is the heat loss through the side wall and at high flow rates the main factor is heat loss with outgoing gases, which is more

pronounced for small lengths of the reactor and for large particle sizes. Another specific feature of the system considered is the intersection of the LCL curves built for short and long reactors. This specific behavior is connected with the formation of a steeper front of the combustion wave due to phase transition and qualitatively discussed above.

At large lengths of the reactor and small sizes of particles, this factor manifests itself weakly, resulting in the intersection of the curves for different lengths of the reactor and different sizes of particles.

The lowest value of the minimum concentration of acetone in water, which were obtained in the calculations are: for the adiabatic case – 13.5% (at the specific liquid flow 0.05 kg/( $\rm m^2$  s) and standard values of other parameters) for the nonadiabatic case – 13.9% (at the specific liquid flow 0.85 kg/( $\rm m^2$  s) and standard values of other parameters). A comparison with the values obtained in [8] shows that in the adiabatic case, the lean combustibility limit (minimum concentration) for a reverse flow reactor is higher compared to a "tube-in-tube" reactor. In the case of typical heat loss parameters the concentration limit for the reverse flow reactor is lower.

The influence of the flow reversing period on the lean combustibility limit for two-phase combustibles is an important problem for further investigation.

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