Self-organization of processes in gas and liquid-phase catalytic reactors

N A Merentsov¹, A V Persidskiy², V V Groshev¹, V A Kozlovtev¹ and A B Golovanchikov¹

¹Volgograd State Technical University, Lenina 28, Volgograd 400005, Russia
²JSC Federal Scientific and Production Centre «Titan - Barricady», Volgograd 400071, Russia

¹E-mail: steeple@mail.ru

Abstract. The paper provides a system of flexible self-organization of hydrodynamic, thermal and diffusion processes in gas and liquid-phase catalytic reactors the purpose of which is to self-adapt the system to the most qualitative chemical catalytic and gas-liquid reactions and search for the best energy transfer modes (resonant modes). The developed flexible system of self-organization of processes in catalytic and gas-liquid reactors implies using elastically deformable layers of catalysts, both independently made from the required materials (contributing to resonance and effective chemical reactions) and applied on blocks of compressed metal shavings with volumetric-elastic properties. This allows affecting the chemical reactions significantly and also solving a very serious environmental problem of recycling manufacturing wastes since various steel grades being mechanically processed at machine-building enterprises form a huge amount of wastes with various properties and configuration; those blocks can undoubtedly be applied as elastically deformable catalyst units in manufacturing.

1. Introduction
In chemical technological systems the attention is focused on the plants for carrying out chemical processes. Let us dwell on the chemical reactors having a wide range of design and mode features which depend on the conditions of the reactions, the rheological characteristics of the reaction masses, the variety of chemical reactions and the methods of influence on the reactions [1-5]. Displacement reactors are plants with an elongated body (channel) providing directed motion and mutual mixing of the reaction mass. There are many traditional factors and actuators to the process such as the hydrodynamic situation in the reactor [6-13], the thermal conditions of the chemical process, the conditions of heat exchange, the presence of a catalyst, the way the process is organized, the nature of the process changing over time, the design features of the plant, etc. One of the most important and complex tasks related to high-quality operation of chemical reactors is to provide the required parameters on industrial scale, since the slightest change of scale and other types of influence (electromagnetic radiation, flow patterns, circulation of heat flows, etc.) immediately fundamentally changes the conditions of the reaction behavior and inevitably affects the parameters of the final reaction products [14-30].
Currently, one of the main tasks is the development of systems for carrying out chemical reactions that have the ability of flexible self-organization that means self-adjusting at the stage of commissioning work, and also during the work, to the reference modes and performance parameters [31-33]. Especially creating (maintaining) of optimal, in particular, resonance working conditions. Self-organization of processes in chemical catalytic and gas-liquid catalytic reactors can be carried out, in the authors’ opinion, with the use of several methods. One of them will be described in detail in this paper. The main idea of self-regulation of reactor systems is the automatic searching (creating) and maintaining of the resonant conditions for the reactions that means creating conditions under which energy is transmitted most accurately that means the self-organization of systems enables to control reactions and the conversion degree, as well as the yield of the target product. A very important feature of the developed system is the ability to carry out successive transformations of the initial materials within one combined reactor consisting of a sequential set of sections with volume deformable catalyst layers (figure 1).

For example, manufacturing of catalyst layers in the form of metal shavings, of certain geometric configurations or applying of catalysts on the surface of the compressed layers of metalworking machine wastes allows sharp developing the contact surface between the reaction mass and the catalyst layer, developing advanced circulation schemes and active mutual mixing of gas and liquid reactions products; and it also enables to actively influence the chemical reactions behavior smoothly changing and adjusting to the required characteristics or impulsively affect the volume of the catalyst which in turn will actively disperse and mix the reaction products at the macro level giving a boost to the chemical reaction due to the resonance effect at the micro level. Not to mention the fact that the use of such materials as elastically deformable sectional layers of catalyst is the way to utilize industrial wastes which appear in large quantities at machine-building and metal-working enterprises [33-41]. This factor will have a significant environmental and technical and economic effect from the point of view of energy and resource saving. But we should not forget that not only wastes from metalworking machines can be used as elastically deformable packed catalyst blocks; with this method (by metal cutting) we can produce a catalyst that will have certain desired geometrical and surface configurations and an accented chemical composition to give the best impulse to the chemical reaction [42-44].

2. Methods and materials
The authors offer to carry out the self-organization of processes in chemical gas-liquid, bubbling and catalytic gas and liquid-phase reactors using smoothly and (or) impulse-deformable layers of catalysts. The reactor is to consist of a sequential set of standardized sections (shells) each of which has a layer of an elastically deformable dispersing catalyst and is packed into a heat-exchanging jacket to control the thermal conditions of chemical reactors (figure 1). Elastically deformable catalyst blocks can be smoothly or impulsively deformed with the help of hydraulic cylinders and the parameters of regulation and the frequency of impulse deformation can vary both at the stage of commissioning work and in the current work of an industrial reactor. The main purpose of flexible self-regulation of hydrodynamic, thermal and diffusion processes is to achieve and maintain the most developed and qualitative characteristics and parameters of chemical reactors performance since during the transition from the laboratory to industrial systems the conditions for the implementation of chemical reactions change drastically and we should have effective tools for influencing and regulating all parameters of the system flexibly.
Figure 1. Sectional gas-liquid bubbling (or catalytic) reactor with a self-organizing process system.

Figure 1 shows a configuration of a bubbling gas-liquid or catalytic reactor for implementing gas and liquid phase reactions; the reactor consists of a sequential set of standardized sections 1; each section has its own configuration specified by the flow patterns of liquid and gas-phase reaction products. Chemical reaction catalysts are applied to the surface of porous and elastic packing materials 2 which are amenable to volume elastic deformation; or the adjustable packing materials themselves are accentuated catalysts and give impulse to chemical reactions. Smooth or pulsed volumetric deformation is carried out under the influence of hydraulic cylinders 3, moving the rigid movable grid 4 limiting the volume of the catalyst layer. The cylinders of each subsequent unified section stop at the rigid fixed grid of the previous section 5. When the catalyst layer is compressed the section’s hydraulic resistance increases, the porosity decreases, the specific surface increases, the channels for flowing reaction products change, the contribution of the viscosity components increases, the flow pattern of the reaction mass changes; as we can see, the conditions for implementing chemical reactions change very substantially. Each of the unified sections contains a heat exchange jacket 6 which we can use for bringing to or taking heat from the products of chemical reactions monitoring the thermal conditions. It should be noted that in case of using elastically deformable metal shavings as catalyst layers the section of the packing material will have adjustable longitudinal heat conductivity in the catalyst material which is to be used and this should be taken into account during engineering and heat calculations.

In column reactors of the proposed design it is possible to carry out high quality sequential (cascade) gas and liquid catalytic reactions. Flexible self-regulation of hydrodynamic, thermal and diffusion processes of the reactor can be carried out autonomously within each individual section or in a centralized way with the simultaneous involvement of all sections in the process of self-organization.

The main task of a self-organizing process system in a catalytic reactor is to identify and maintain optimal operating working modes corresponding to the given inversion degree of the reaction mass and to search for resonant effects of chemical reactions. Figure 2 shows the configuration of a multisection catalytic reactor with the flexible system of self-organization processes.
Figure 2. Scheme of the self-organization system of a gas-liquid bubbling or (gas) liquid-phase catalytic reactor.
Using the example of section 1, each section of the reactor consists of an elastically deformable layer of catalyst laid between two grids. The upper (lower) grid $GU$ ($GL$) is driven by the hydraulic cylinders $HC1.1$ and $HC1.2$. Due to the oil pressure, the hydraulic cylinders lower the grid and squeeze the packing. The grid is returned to the upper position due to the injection of oil into the lower chambers of the cylinders and the elasticity of the catalytic layer. The ultrasonic sensor $S1.2$ mounted on the body of one of the hydraulic cylinders identifies the distance to the target $CT$ which is a metal plate mounted on a bracket on the upper grid, thus the position of the grid can be identified by the readings of the $S1.2$ sensor. Below the elastically deformable catalyst layer the sensor $S1.3$ is installed, it identifies the concentration of the reaction products. The type and design of this sensor depend on the properties of the reaction products and technological requirements. An oil flow rate sensor $S1.1$ is installed in the hydraulic line supplying oil to the upper chambers of the hydraulic cylinders. All the sensors transmit digital data to a programmable logic controller $PLC$. The controller handles the hydraulic valves by closing the discrete contacts and giving digital commands and the valves $V1.2$, $V1.3$, $V1.4$, $V1.5$ have an electromagnetic drive and are driven by discrete signals, and valve $V1.1$ is driven by a stepper motor and allows controlling the flow area of the hydraulic line. The valves $V1.2$ and $V1.3$ handle the injection of oil into the upper and lower chambers of the hydraulic cylinders, respectively, and $V4$ and $V5$ are used for dumping oil from the cylinder chambers into the oil reservoir $OR$. Oil pressure is provided by the oil pump $P$ driven by an electric motor $M$. The hydraulic batteries $B1.1$ and $B1.2$ smooth out pressure surges in the line when valves $V1.2$ and $V1.3$ are open, and backpressure valves $V1.1$ and $V1.2$ prevent oil from moving towards the pump.

![Figure 3. Algorithm of processes self-organization in a gas-liquid or catalytic reactor.](image-url)
The common elements for the three sections here are the oil pump $P$ with the electric drive $ED$ and the programmable logic controller $PLC$ which collects data from 9 sensors and handles 15 valves 3 of which are smoothly driven by a stepper motor.

Figure 3 shows the algorithm of the control program which is cyclically executed in the $PLC$. Simultaneously for each of the sections it estimates the concentration at the exit of the catalyst layer and specifies the operation mode of the elastic-deformable catalyst layer, and its parameters, which provides the given concentration of reaction products at the output of the corresponding section.

Using the example of section 3 the program operates as follows. Before starting the extractor the operator enters the following parameters: amplitude $A$ of the catalytic layers pulsation as a percentage of the maximum possible, the initial pulsation frequency $F_{0_1} \ldots F_{0_3}$ with which automatic selection of the optimum frequency will begin, step of the frequency adjustment $N$ determining the accuracy of the optimal pulsation frequency, the required concentration of the catalysis product at the exit of each of the packings $C_{opt1} \ldots C_{opt3}$ and the determination accuracy of the concentration $Acc$ which specifies the maximum allowable deviation of the concentration from the optimum.

At the next stage, the program loads the values of the working volume of the hydraulic system of each section $V1 \ldots V3$, the lower $H_{0_1} \ldots H_{0_3}$ and upper $H_{max_1} \ldots H_{max_3}$ grid coordinates from the controller memory. These values are specified during the section calibration process. Then the program assigns the initial values to the variable storing the value of the optimal frequency, as well as to the variable $Pulse1 \ldots Pulse3$ which determines whether the operation mode of the packing is going to be static or pulsating.

Then the program specifies, depending on the parameters set by the operator, the lower limit that the grid should reach in the pulsating mode; then it specifies the step of the grid position change in the static mode.

The program receives from sensor $S3.3$ the value of the actual concentration of the reaction product at the exit of the packing, and then calculates the lower and upper limits of permissible concentration.

Next, the program checks whether the actual concentration of the catalysis product is less than the minimum allowable. If it is less, the program checks in which mode the reactor is currently operating. If the value of the variable $Pulse3$ is equal to 0, the mode is static; if it is equal to 1, it is pulsation. In the static mode, the grid constantly compresses the catalyst layer to a level corresponding to a given concentration of the catalysis product at the exit of the section. In the pulsating mode the grid reciprocates with a given amplitude and frequency corresponding to a given final concentration of the reaction mass (degree of conversion).

In the static mode the controller gives a command to open valve $V1$ by half of its capacity which provides smooth moving of the grid during regulation, and then by one step increases the value of the variable $Lvlopt_3$ which specifies the optimal position of the grid providing the desired product concentration, i.e. initiates its lowering by one step.

Then the program checks whether the value of the optimal position of the grid has reached the lower limit of the grid mobility $H_{max_3}$. If it has, the program switches the reactor operating mode to pulsation.

If the operation mode is pulsating, the program increases the value of the pulsation frequency of the packing $F_{opt_3}$ by one step.

If the actual concentration of the catalysis product $Sfakt_3$ is greater than the permissible $C_{max_3}$, the program in the static operation mode reduces the value of the variable specifying the optimal position of the grid $F_{opt_3}$, i.e. initiates its rise by one step. In case of a pulsating mode the program reduces the pulsation frequency of the packing $F_{opt_3}$ by one step and then checks whether the specified pulsation frequency is less than the minimum allowed. If it is, the program switches the reactor to a static mode.

After that, the execution of the algorithm branch described above is interrupted for 5 minutes.

Parallelly with those actions, an algorithm that organizes the deformation of the catalyst layer in accordance with a specific mode is performed.
The program checks which operation mode of the reactor is set up. In the static mode the program receives the current grid coordinate $Pos3$ from the sensor $S3.2$, and then compares it with the coordinate value $Lvlopt_3$ calculated earlier. If the value of $Pos3$ is less than $Lvlopt_3$, the grid is above the optimal position, and the catalyst layer must be compressed. The controller gives a command to close the valves $V3.3$ and $V3.4$, and then to open the valves $V3.2$ and $V3.5$. This causes oil to be pumped into the upper chambers of the hydraulic cylinders and the grid is lowered and the catalyst layer is compressed. If the value of $Pos3$ is greater than $Lvlopt_3$, the grid is below the optimal position, and the catalytic layer must be decompressed. The controller gives a command to close valves $V3.2$ and $V3.5$, and then to open valves $V3.3$ and $V3.4$. The grid rises, and the catalyst layer is decompressed. If the value of $Pos3$ is equal to $Lvlopt_3$, all valves are closed, the grid is stopped and fixed.

In the pulsation mode, the program reads the current grid coordinate from sensor $S3.2$. If the grid coordinate is less than the required one, the controller gives a command to close the valves $V3.3$ and $V3.4$ and open the valves $V3.2$ and $V3.5$. The oil is injected into the upper chambers of the cylinders and dumped from the lower ones and the grid begins to move downward compressing the catalyst layer. If the grid coordinate is greater than the required one, the controller gives a command to close valves $V3.2$ and $V3.5$ and open $V3.3$ and $V3.4$. The oil begins to be pumped into the lower chambers of the cylinders and dumped from the upper ones and the grid begins to move upwards.

Next, the program calculates what the oil consumption in the line should be when it is pumped into the upper chambers of the hydraulic cylinders and dumped from them, gets the actual oil consumption data from $S3.1$ sensor and compares them with the required value. If the actual oil consumption is less than the required one, the controller commands the valve drive $V3.1$ to open it by one step, if it is greater, the controller commands to close it by one step. If it is equal to the required one, it does nothing. As a result, the more valve $V3.1$ is open, the faster the oil fills the hydraulic cylinders and is dumped from them, hence the frequency of cyclic movements of the grid becomes higher. When the grid reaches the top position, valves $V3.3$ and $V3.4$ close.

Thus, when cyclically performing this algorithm, the grid moves down and up causing pulsation of the elastically deformable catalyst layer.

In parallel, the program receives from the sensor $S3.3$ the value of the actual concentration of the extracted substance and compares it to the required value. If the concentration is less than the set concentration, the program increases the frequency of catalyst layer pulsations by step $N$, if the concentration is greater, it decreases the frequency of catalyst layer pulsations by the same step.

The values of the working volume variables $VX$, the lower $Hmax_X$, the upper $H0_X$ extreme coordinates of the grid depend on the geometrical dimensions and design of the specific plant. They are specified during the calibration of the reactor in automatic mode. Figure 4 shows the calibration algorithm for a multisection catalytic reactor in automatic mode.

After launching, the program gives a command to open valve $V1.1$ to a maximum, then close valves $V1.2$ and $V1.5$ and open $V1.3$ and $V1.4$. The grid of section 1 begins to rise to its highest position. Then the program receives from the $S1.1$ sensor the current value of the volume oil consumption and checks whether it is equal to zero. The program repeatedly receives the data from the sensor and checks the volumetric oil consumption until its value becomes equal to 0. The zero movement of oil in the hydraulic line means that the grid has risen up to the stop and is not moving. Then the program receives the distance to the target from the sensor $S1.2$ and writes it into the variable $H0_1$ as the initial coordinate of the grid. Then the program closes valves $V1.3$ and $V1.4$ and opens valves $V1.2$ and $V1.5$, after that it starts counting the time in seconds using the system timer. The program assigns zero values to the variable-counters: $Cnt$, counting the number of cycle passes and $EQ1$, summing the value of the actual volume flow rate of oil obtained from sensor $S1.1$ during each cycle with the previously obtained values.

Next, the program sums the values of the actual volumetric oil consumption and the number of cycle passes. After that, it checks whether the volume flow of oil is zero. If it is equal, then the catalyst layer is maximally compressed and the grid no longer lowers.
Figure 4. Calibration algorithm of a self-organizing system parameters of a catalytic reactor.
As soon as the volume flow rate of the oil is zero, the program stops the time counting and fixes its value in the variable t, and then in the next step, using the formula that includes the average volume flow rate of oil for the entire time of the grid lowering and the time of its lowering; it calculates the oil volume pumped into hydraulic cylinders with a maximum compressed catalyst layer $V_1$. Then the program receives the distance to the target from the $S1.1$ sensor and writes it into the variable $H_{max \_1}$ as the grid coordinate corresponding to the maximum compressed elastically deformed catalytic unit.

The program then stores the obtained $V_1$, $H_{max \_1}$ and $H_{0 \_1}$ extreme grid coordinates in the PLC permanent memory, gives a command to close valves $V1.3$ and $V1.4$ and open valves $V1.2$ and $V1.5$ to feed the oil into the lower sections of the hydraulic cylinders and decompress the packing. The program then proceeds to calibrate the subsequent sections (2 and 3) in the same way after which the calibration process is completed.

It should be noted that the system of flexible processes self-organization gives unique possibilities for the current monitoring of the performance of chemical reactors. Static control modes can be applied in gas-phase catalytic reactions, and pulsed pulsation modes can be applied in liquid-phase catalytic reactions, despite the fact that the oscillation energy of elastically deformable catalyst layers will be transmitted to liquid-phase reaction products throughout the volume resulting in increased mutual mixing of reaction products and active dispersion liquid phase products.

3. Conclusions

The authors have considered a new direction of chemical engineering that allows adapting the system of processes occurring in the chemical reactor to the reference parameters (modes) efficiently and directly during the work; and in some cases the given direction allows catching resonant effects at the micro level, i.e. creating conditions for flow patterns and mutual mixing of reaction products that allow chemical transformations in the conditions of the highest energy transmitting (resonance). Not to mention the fact that such self-organizing systems allow catalytic reactions to be carried out in a high-frequency vibration mode due to volumetric impulsive elastic deformation, and the energy will be transmitted to the reaction mass from the catalyst layer having ribbed sharp (cutting) edges, which results in the highest quality mutual mixing and dispersion of the reaction products which is especially important for liquid-phase reaction products.

This new direction of chemical engineering opens up unique opportunities for quality current operational impact on the process, especially the so-called fast reactions. Such self-organization of hydrodynamic, thermal and diffusion processes in gas and liquid-phase bubbling and catalytic reactors allows the systems to automatically adjust to the reference operation modes and using the programmable controllers and automation tools make the developed systems incredibly flexible and intelligently adaptable. Moreover, self-organization of all processes can be achieved by the system in the search mode analyzing the conversion degree (conversion) of the reaction mass.

We should also note the design features of the developed reactor with flexible processes self-organization from the point of view of controlling the conversion degree; also having a unique ability to perform cascade sequential transformations of the initial materials within one reactor, each section being automatically regulated and packed into a heat-exchanging jacket, i.e. the flow patterns of reaction masses and the heat modes within each section are underaccented control.

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