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Theoretical and Experimental Aspects of Using Structured Catalytic Systems¹

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Abstract—Mass transfer in honeycomb monolith catalysts has been theoretically and experimentally studied by methods of computational hydrodynamics and NMR tomography. Theoretical estimates have been obtained for optimum parameters of processes employing honeycomb monolith catalysts. Newly developed catalysts for selective ammonia oxidation have provided a technological basis for nitrous oxide production with a selectivity of 85–88%, and a pilot plant using these catalysts has been designed. Methods of mathematical description have been developed for catalytic processes in fibrous materials, and the potential of these materials in soot aerosol removal from waste gas has been demonstrated. Principles of structuring of granular catalyst beds have been developed through theoretical analysis and experimentation. Results of commercial-scale application of structured beds in tubular and adiabatic reactors are presented.

The efficiency of new catalysts in the vast majority of cases is determined by process design. The challenge is, first of all, to design a catalytic reactor adequate to the properties of the catalyst. A convincing example demonstrating the validity of this statement is catalytic cracking of vacuum gas oil. At virtually the same activity of the catalyst, replacement of a fluidized-bed reactor by a gas-lift reactor decreases the catalyst charge and, accordingly, increases the reactor capacity by a factor of about 10. Thus, the search for a new catalytic process design is commercially very promising. The catalysts most widely used at the moment are produced as particles of different sizes and shapes: spheres, sticks, rings, fine powder, etc. Unfortunately, catalyst optimization can improve a process only to a limited extent. In most cases, the efficiency of the most active catalysts in real industrial reactors is limited by heat and mass transfer.

In recent years, catalysts of new geometric shapes, e.g., honeycomb monolith catalysts [1–3] and fibrous catalysts [4, 5], have been actively designed. In some cases, catalytic processes can be intensified by structural organization of the catalytic reactor using fixed catalyst beds allowing dispersed-phase filtration [6, 7], special methods of loading [8], and turbulent structures in fluidized beds [9, 10]. What all of the above process designs have in common is the presence of some structures, either determined by the catalyst shape (e.g., monolith or fibers) or formed directly within the catalytic reactor. This paper deals with the development of

theoretical (including mathematical) principles of using the above structured catalytic systems.

The purpose of this study is to form a theoretical basis for designing catalytic reactors operating primarily at short contact times (on the order of 10^{-2} – 10^{-3} s) and to develop catalysts for such reactors.

Our study included the following:

- (1) theoretical and experimental investigation of heat and mass transfer in structured systems,
- (2) development of methods for mathematical modeling and optimization of processes at millisecond contact times,
- (3) development of mathematical methods and experimental techniques for controlling granular structures in catalytic reactors, and
- (4) investigation of catalytic properties and design of catalysts for structured systems.

THEORETICAL AND EXPERIMENTAL PRINCIPLES OF HEAT AND MASS TRANSFER IN HONEYCOMB CATALYSTS

The flow of a reactive gaseous medium filtering through channels of a honeycomb monolith catalyst is usually laminar. Only at the inlet of the catalyst bed is the flow unsteady, and it is generally assumed that the mass-transfer coefficients in this region significantly exceed their steady-state values [1]. The validity of these assumptions was tested experimentally by NMR tomography [11] and theoretically by methods of computational hydrodynamics.

The results of NMR tomographic experiments are presented in Fig. 1, which also shows a schematic of the experimental setup used. The experiment consisted in measuring the proton density (water content) of a

¹ The articles by A.S. Noskov and by A.V. Vorontsov *et al.* continue a series of publications dedicated to the 45th anniversary of the Boriskov Institute of Catalysis, Siberian Division, Russian Academy of Sciences. Earlier, we published a special issue of the journal dedicated to this event (2005, no. 2).

monolith catalyst as a function of time. A catalyst particle was presaturated with moisture and then blown with dry air. Figure 1b plots the tomographic signal versus time and monolith length. The measurements were made at points spaced by 2.5 mm along the monolith length. Figure 1c shows the profile of moisture evaporation rate along the honeycomb monolith length at different filtration velocities (Reynolds numbers $Re = \frac{Ud}{\nu}$, where U is the gas velocity in the channel, d is the linear size of the channel, and ν is gas viscosity). It can be seen that the evaporation rate decreases with increasing distance from the monolith inlet. The length of the channel region in which the flow is unsteady reaches almost half the monolith length, which considerably exceeds published data [1].

To theoretically explain the experimental results obtained, gas flow was modeled by solving the Navier–Stokes equation using the Fluent software (Fig. 2). Among the calculation results obtained, note the following two facts. Up to about half the monolith length (12 mm), the gas flow is unsteady because of eddies (Fig. 2b). It is the eddies at the inlet of the honeycomb monolith that cause the observed increase in the mass-transfer coefficients and, hence, the evaporation rate.

Thus, the following specific features of catalytic processes in honeycomb catalysts should be taken into account in process design:

The heat- and mass-transfer coefficients vary along the honeycomb monolith.

There is no conductive heat transfer along the length of the monolith because the monolith has an insignificant thermal conductivity.

As a rule, the reaction is controlled by external mass transfer.

By way of example, Fig. 3 shows how methanol conversion in air cleaning depends on the geometry of a bed packed with honeycomb monoliths. Two cases were considered in our calculations, namely, a process performed on a single monolith 150 mm in length and a reaction occurring on two 75-mm-long monoliths (the total length is again 150 mm). Since the reaction is controlled by external mass transfer, the monolith divided into two parts allows one to use the effect of eddies at the inlets of the parts, owing to which conversion increases from 81% (on a single 150-mm-long monolith) to 98% (on two 75-mm-long monoliths). Along with conversion, the maximal temperature also increases (from 323 to 374°C). However, a further increase in the number of monoliths to three or four increases methanol conversion insignificantly (only by 1.5–1.7%).

In most cases, honeycomb catalysts contain precious metals, e.g., platinum or palladium. Therefore, it is desirable to reduce the precious metal content without impairing the process. Figure 4 presents the profiles of the maximum possible mass-transfer and reaction

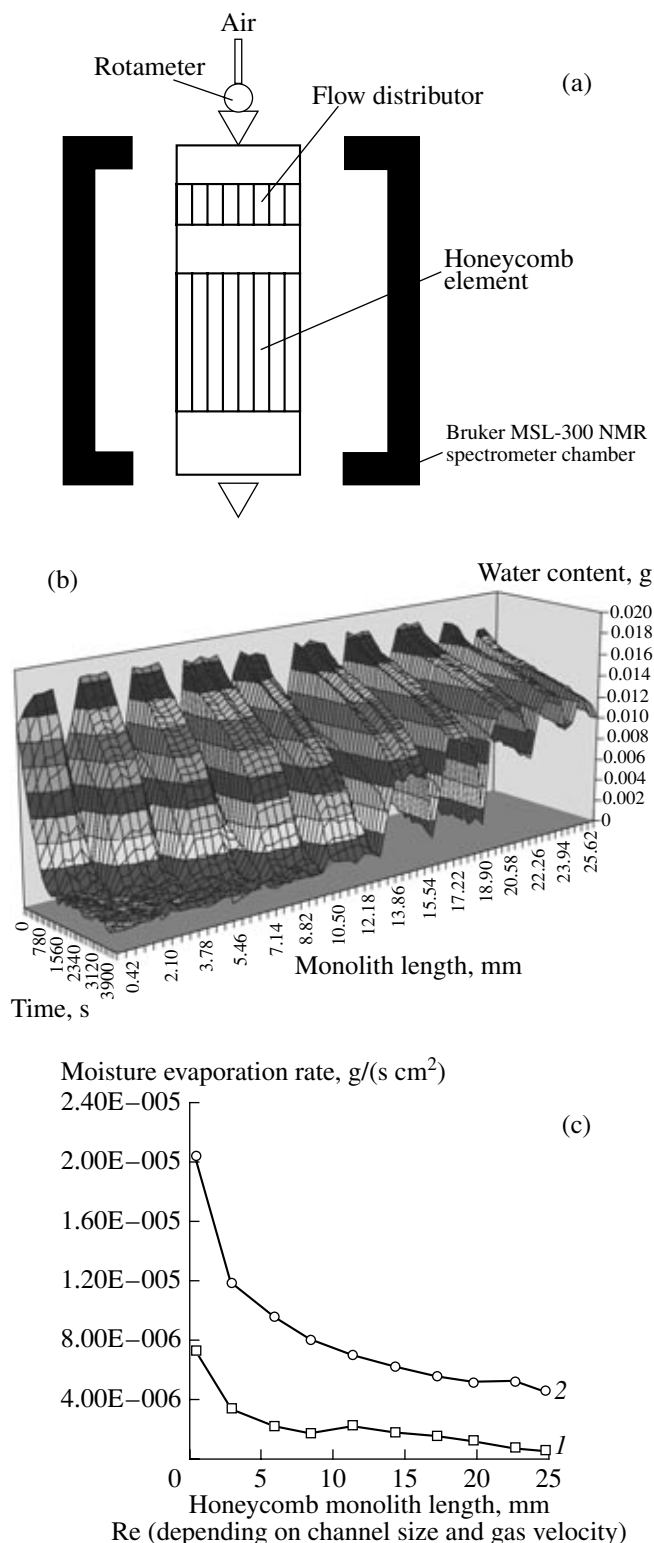


Fig. 1. (a) Application of NMR tomography to determining the local mass-transfer characteristics in honeycomb monolith catalysts at $Re = (1)$ 16 and (2) 195, (b) tomographic signal versus time and monolith length, and (c) profile of moisture evaporation rate along the honeycomb monolith length at different filtration rates (Re values).

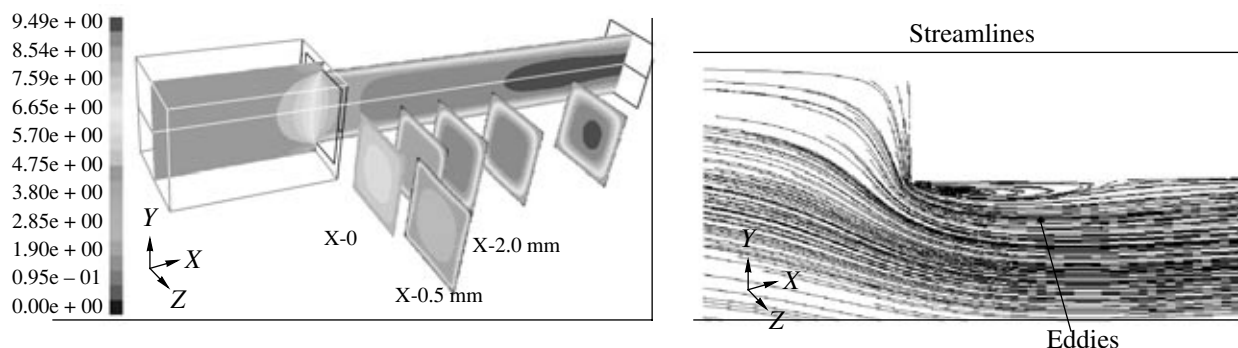


Fig. 2. Modeling of the gas flow pattern in a honeycomb monolith catalyst by solving the Navier–Stokes equation.

rates along the catalyst bed (honeycomb monolith) length. At the inlet of the bed (region A), the mass-transfer rate is maximal and the total conversion is limited by the insufficiently high reaction rate. Downstream (region B), the reaction is limited by mass transfer; i.e., in this region, a high catalyst activity is unnecessary. In region C, the reaction rate is a limiting factor again. Such variation of reaction and mass-transfer rates along the bed length poses the problem of optimizing the active component distribution over the honeycomb catalyst bed. There can be two different formulations of this problem [12]:

(1) Determination of the minimal active-component content (M) of the honeycomb monolith that is necessary to attain a given conversion.

(2) Determination of the maximal outlet conversion at a fixed weight of the active component.

Theoretically solving these problems required new methods for analyzing mathematical models of catalytic reactors. It is possible to reduce the problem of optimization of the active-component distribution to solving an Euler differential equation of special form [13].

Figure 5a compares calculated conversion profiles along the reactor length for the uniform (curve 1) and optimal (curve 2) active-component distributions for a model reaction (first-order deep oxidation). These active-component distributions are shown in Fig. 5b [12].

The results obtained can be characterized as follows: for irreversible catalytic reactions, optimization

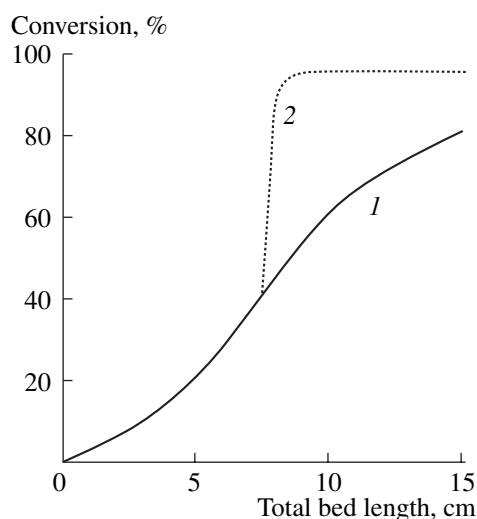


Fig. 3. Calculated methanol conversion versus total length of the catalyst bed ($\text{Pt}/\text{Al}_2\text{O}_3$) at $T_{\text{in}} = 80^\circ\text{C}$, a methanol concentration of 1.2 vol %, $u = 2.0$ m/s, a channel diameter of 2 mm, and a wall thickness of 0.3 mm for (1) a single monolith 150 mm in length ($T_{\text{max}} = 323^\circ\text{C}$) and (2) two 75-mm-long monoliths ($T_{\text{max}} = 374^\circ\text{C}$).

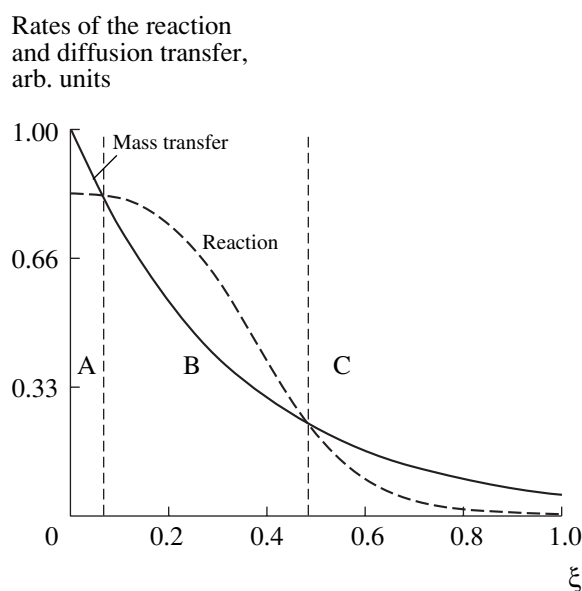


Fig. 4. Profiles of the maximum possible reaction rate and the maximal possible rate of diffusion mass transfer to the catalyst surface along the catalyst bed length ξ .

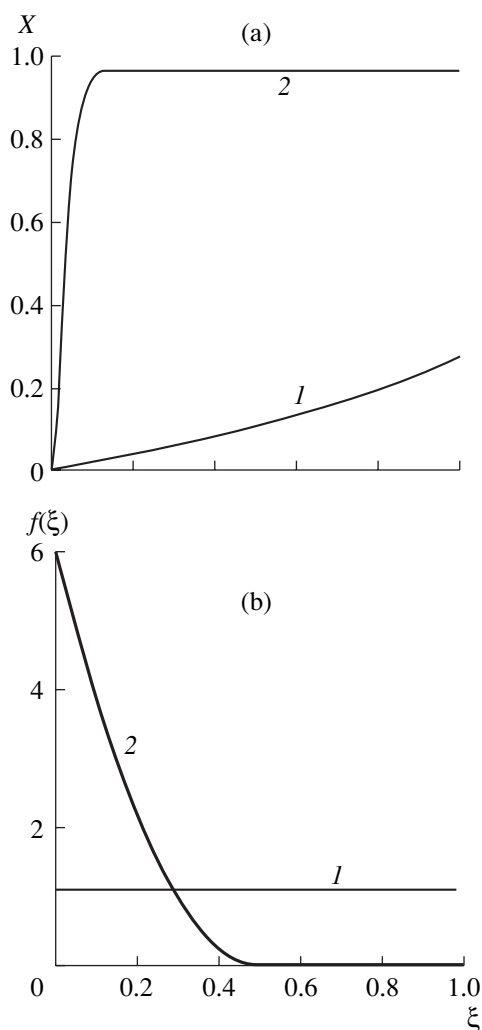


Fig. 5. Optimization of the active-component distribution along the honeycomb monolith catalyst length: (a) conversion profiles along the reactor length for the (1) uniform and (2) optimal active-component distributions; (b) the (1) uniform and (2) optimal distributions of the active component.

of the active-component distribution allows one to reduce the precious metal content by 30–50% at the same conversion [12, 13].

Some of the results obtained were used in the development and commercialization of nonplatinum honeycomb catalysts in the two-step process of ammonia oxidation in nitric acid production [14]. This process is carried out in more than ten UKL-7 apparatuses and enables one to reduce precious metal consumption by 30–40% and the irreparable platinum loss by 25%.

The theoretical and experimental results obtained on honeycomb monolith catalysts can also be used in the development of catalytic microreactors [15]. Catalytic microreactors are monolith catalysts with channels several tens of micrometers in diameter and intensive heat supply (release). Such catalytic reactors are promising

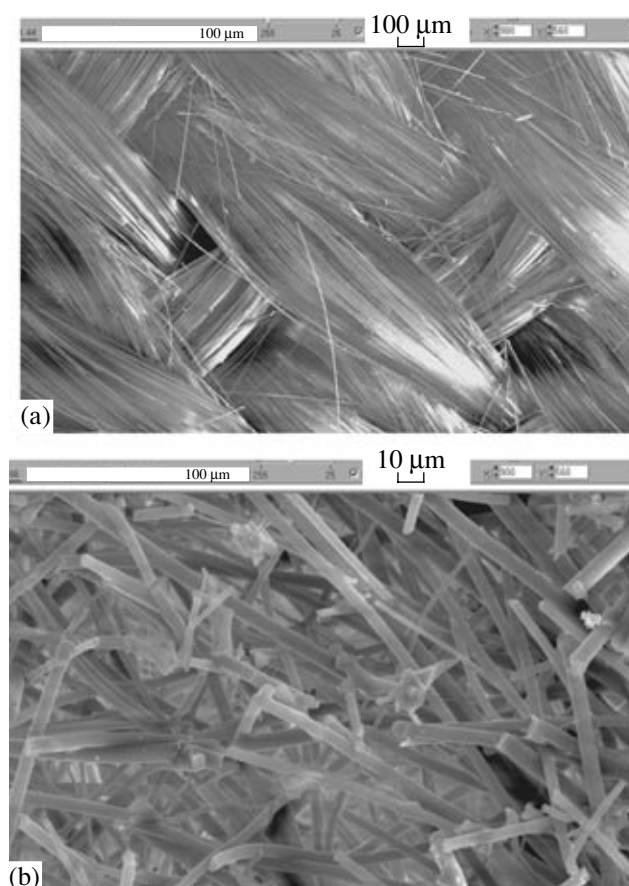


Fig. 6. Catalytic systems based on fibrous material: (a) glass cloth and (b) irregular fibrous quartz.

owing to their very high output capacity per unit volume. Suffice it to say that an ammonia oxidation microreactor 1 cm³ in volume can produce up to 10 t of NO per annum. Catalytic microreactors are most promising for hydrogen production in vehicle-mounted fuel cells and for selective small-scale processes.

CATALYTIC SYSTEMS BASED ON FIBROUS MATERIALS

Catalysts based on fibrous materials are currently being actively designed [5]. These materials exhibit unique catalytic properties in a number of reactions. Known fibrous materials are glass cloth [5] (Fig. 6a) and irregular fibrous quartz (Fig. 6b).

To evaluate the hydrodynamics of gas flows in glass cloth systems in a two-dimensional approximation, Klenov performed calculations using the Navier–Stokes equation. For a gas velocity of 0.1 m/s and a filament spacing of 10 μm, the calculations demonstrated that the mass-transfer coefficients on individual filaments within a fiber are negligibly small and only the mass-transfer coefficients on outer filaments are rather large. These specific features of mass transfer require a

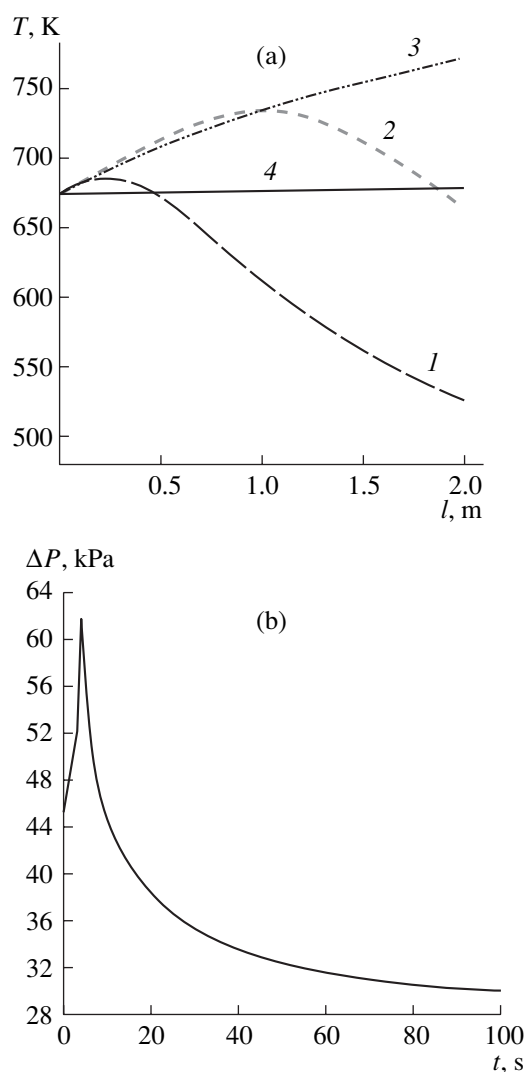


Fig. 7. Calculated regime of oxidative regeneration of a fibrous-quartz catalytic soot filter: (a) temperature profile along the filter length l at $t = (1)$ 2, (2) 4, (3) 10, and (4) 200 s and (b) evolution of the pressure drop across the filter during regeneration.

special design of glass-cloth catalytic reactors. Mass transfer in glass-cloth systems can be explored in more detail using modern physical methods (e.g., NMR tomography) and computational hydrodynamics applied to a three-dimensional model of glass cloth.

Materials based on fibrous quartz are promising for catalytically active filters for collecting and oxidizing soot particles in diesel exhaust. Catalysts that accelerate the oxidation of solid soot particles are currently being actively developed. For example, Ivanova and colleagues (Boreskov Institute of Catalysis, Siberian Branch, Russian Academy of Sciences, Novosibirsk, Russia) have designed oxide catalysts capable of decreasing the ignition point of model soot (so-called diesel ash) from 435°C (without a catalyst) to 200–250°C (in contact with a catalyst). However, along with

a catalyst, it is necessary to design a filter to remove soot particles from exhaust gas. Considering the typical size of soot particles, such a filter can be made from materials based on fibrous quartz (Fig. 6b).

For theoretical analysis, we constructed a mathematical model for collection of soot aerosol from diesel exhaust by fibrous material. This model includes unsteady-state equations for the concentrations of soot particles, O_2 , and CO_2 in the gas phase; the soot content of the solid phase; and filter temperature. Furthermore, it includes an equation for the pressure drop across the filter [16]. The main results are obtained by modeling the dynamic regimes of soot collection and the oxidative regeneration of a fibrous filter supporting a catalyst. Calculations showed that soot collection efficiency increases as the filter is filled with soot. The thermal conditions are determined under which the filter will only be filled with soot and under which the filter will be filled with soot and simultaneously oxidized and self-regenerated. In the former case, the filling of the filter with soot is accompanied by an increase in the pressure drop up to a limiting value of 0.5 atm. In the latter case, the pressure drop rises insignificantly to reach an asymptotic value depending on the temperature of the soot-containing gas.

Figure 7 presents calculated data showing how the temperature profile along a catalytic reactor changes as the temperature of the feed gas is increased from 473 to 673 K (Fig. 7a) and how the pressure drop changes during oxidative regeneration (Fig. 7b). As the feed temperature is raised to 673 K, the soot collected in the filter starts to oxidize and the temperature begins to grow (Fig. 7a). Because of the increase in temperature, the pressure drop across the filter increases, reaching a maximum 8–10 s after the beginning of regeneration. Subsequently, as soot burns out, the pressure drop decreases, reaching an asymptotic value.

The calculations performed allowed us to estimate the main parameters of fibrous-quartz catalytic soot filters showing 80–85% efficiency.

TURBULENT FLUIDIZED CATALYST BED

One of the main problems in using a fluidized catalyst bed is the formation of gas bubbles, which decreases conversion and complicates scaling up from laboratory reactors to pilot reactors.

The motion of a gas bubble in a fluidized catalyst bed gives rise to a so-called wake. If the bed is fluidized in the laminar (bubble) regime, small bubbles entering the wake remain stable and coalesce with a large bubble, whose size is thus increased. If the bed is fluidized in the turbulent regime, bubbles entering the wake collapse and do not grow. In this case, the fluidized bed is characterized by large gas–catalyst mass-transfer coefficients [9].

For a turbulent fluidized bed to exist, a number of conditions should be met [9]:

(1) Gas velocity in the reactor should be high enough to ensure a Re value sufficiently large (normally 100–600) for the formation of a turbulent wake layer and to ensure that the fraction of gas bubbles in the bed is $\epsilon = 0.25$ – 0.30 , favoring intensive interaction.

(2) Particles of fluidized material should be light enough since, for $Ar > 100$ ($Ar = \rho_g g d_p^3 / (\rho_p - \rho_g) \mu^2$ is the Archimedes number, where ρ_g is gas density, ρ_p is the density of fluidized particles, d_p is the mean diameter of fluidized particles, μ is gas viscosity, and g is the acceleration of gravity), a transition from bubble flow to pneumatic transport takes place as gas velocity is increased.

A turbulent fluidized bed is characterized by a small bubble size, high interfacial mass-transfer rates, and low amplitudes of pressure fluctuations in the bed because of the small size and instability of gas bubbles.

The properties of turbulent fluidized beds were utilized in the development of a new large-scale process for nitrous oxide production by selective ammonia oxidation [10, 17]. To design this process, it was necessary to develop supported manganese–bismuth oxide catalysts [18]. Such a catalyst of composition MnO_2 – $Bi_2O_3/\alpha-Al_2O_3$ shows a high N_2O selectivity (up to 90%) over a wide range of experimental conditions, producing only small amounts of NO . It was experimentally found that the overall ammonia oxidation rate increases with increasing oxygen and ammonia concentrations and decreases with increasing water concentration. Nitrous oxide selectivity increases with increasing oxygen and water concentrations and decreases with increasing ammonia concentration. Thus, the maximal selectivity with respect to nitrous oxide is reached at high oxygen and water concentrations and a low ammonia concentration. When oxygen is in excess, nitrous oxide selectivity is as high as 90%. Note that, even when the ammonia-to-oxygen ratio is stoichiometric, nitrous oxide selectivity is only a little smaller (88%). Throughout the temperature and composition ranges studied, nitrous oxide selectivity increases with increasing ammonia conversion. This can be explained by the fact that ammonia oxidation occurs via two independent routes and, with a decrease in ammonia concentration, nitrogen formation rate decreases more rapidly. Nitrous oxide selectivity to increases as the temperature is raised from 320 to 360°C. The experiments showed that a further increase in temperature is accompanied by a decrease in N_2O selectivity and a gradual increase in NO selectivity. Thus, the highest nitrous oxide selectivity is attained between 340 and 360°C.

These results underlay the development of a technology for nitrous oxide production by ammonia oxidation in a fluidized catalyst bed (MnO_2 – $Bi_2O_3/\alpha-Al_2O_3$). Since nitrous oxide is to be produced in large amounts (up to 10^5 t/year), it seems promising to use a concentrated inlet reaction mixture consisting only of approx-

Calculated characteristics of nitrous oxide production reactors

Characteristic	Bubble regime	Turbulent regime
Reactor capacity, thousands of tons per year	100	100
Reactor diameter, m	5.4	2.5
Fluidized bed height, m	3.6	11.8
Pressure drop across the reactor, atm	0.5	1.2
Ammonia conversion, %	90	98
N_2O selectivity, %	86	86
Bed volume, m^3	73	31

imately equimolar amounts of NH_3 and O_2 (i.e., 50 vol % NH_3 + 50 vol % O_2). The great adiabatic heating at the reactor inlet, which is $\sim 80^\circ C$ per percent of ammonia, requires a fluidized catalyst bed for converting most of the ammonia and oxygen.

The hydrodynamics of the fluidized bed was described using a two-phase model [19] under the following assumptions:

- (1) Gas in the bubble phase moves in the plug-flow regime.
- (2) The longitudinal dispersion of the gas in the dense phase of the catalyst is taken into account.
- (3) The reaction takes place only in the dense phase.
- (4) The bed is isothermal.

Calculation results for reactors operating in the bubble and turbulent fluidization regimes are compared in the table.

The data presented in the table suggest that, in the case of turbulent fluidization, the amount of catalyst can be decreased by a factor of more than 2.5 with a simultaneous increase in the ammonia conversion (from 90 to 98%).

The studies reported here made it possible to design the world's first pilot plant for nitrous oxide production by ammonia oxidation in a fluidized catalyst bed with an annual output of up to 8 t. If a concentrated reaction mixture (50 vol % NH_3 + 50 vol % O_2) is used, the N_2O content of the reaction products is 85 vol % and the residual NO_x content does not exceed 2 ppm.

FORMATION OF STRUCTURES IN FLUIDIZED-BED CATALYTIC REACTORS

What are the requirements that a catalyst bed must meet so as to work long and efficiently and fully exhibit the designed catalytic properties? It is obvious (and very important) that the bed should have a uniform spatial structure such that catalyst particles are equally accessible for the reactants. This is necessary to perform the totality of chemical reactions in the bed under optimal hydrodynamic conditions and to ensure that all

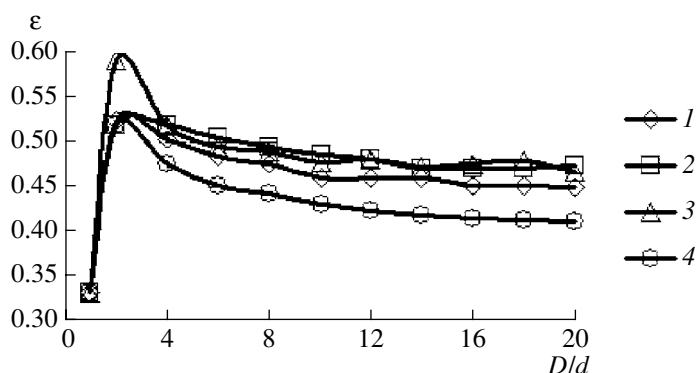


Fig. 8. Granular-bed voidage ε as a function of the ratio of the reactor diameter D to the grain diameter d for catalyst beds in which particles are placed (1) in firm positions beginning at the reactor wall, (2) in firm positions beginning at the reactor axis, (3) in specially (randomly) chosen positions, and (4) in positions minimizing the potential energy.

of the catalyst is engaged in the reaction and is deactivated uniformly.

Another significant condition is that the bed must be durable when used in an industrial reactor. During the process, the bed should not change its volume; i.e., it should show minimal shrinkage under variable dynamic loads, either determined by process conditions or caused by force majeure such as process startups and shutdowns. This requirement is particularly important for radial-flow reactors such as reforming ones.

Studies performed at the Boreskov Institute of Catalysis showed that these problems can be solved by using special techniques for catalyst charging into the reactor.

It was found that a fixed catalyst bed has a “memory,” “memorizing” irregularities and voidage that form as the reactor is charged. To develop a theory of bed structuring, mathematical methods were created to describe the formation of a granular bed [20]. Figure 8 plots bed voidage ε calculated as a function of the ratio of the reactor diameter D to the catalyst grain diameter d (D/d) for the following ways of grain packing:

- (1) Grains are put in firm places beginning at the reactor wall.
- (2) The same is done beginning at the reactor axis.
- (3) Grains are put in specially (randomly) chosen places.
- (4) Grains are put in places minimizing the potential energy.

According to calculated data (Fig. 8), as the reactor size is increased, the densest structure is formed when grains are packed so as to minimize the potential energy and the loosest structure is formed when particles are packed randomly. The experimental finding that bed voidage decreases with an increase in the height from which particles are dropped [8] can be explained by the breakdown of loose structures because of the rapid fall of the grains and by the motion of these grains to places at which the potential energy is minimal.

The results obtained were used to develop catalyst chargers for adiabatic and tubular reactors. To date, the total amount of catalyst charged has exceeded 2000 t (in more than 30 reactors).

On the average, the methods described here allow one to increase the amount of catalyst loaded by 8–12% in comparison with conventional methods.

The totality of new characteristics of granular catalyst beds—uniform structure for filtering flow, increased packing density, and stability under variable loads—increases the reactor capacity (by 5–10%) and catalyst lifetime.

CONCLUSIONS

The field of research presented in this paper can be divided into two parts. The first is development of efficient ways of using catalysts with a regular structure (honeycomb and fibrous catalysts). On the other hand, certain structures (including a turbulent fluidized bed and dense granular structures in fixed-bed reactors) can form in catalytic reactors themselves. Utilization and control of these structures can significantly increase the efficiency of catalytic reactors.

The following has already been done along these lines of investigation:

- (1) NMR tomographic methods have been developed for experimental investigation of mass-transfer processes in structured catalytic systems, and computational hydrodynamics has been used in the theoretical analysis of these processes.
- (2) Mathematical methods for describing the formation and the behavior of structured systems have been devised.
- (3) Catalysts for structured systems have been developed and studied.
- (4) Promising fields of application have been found for structured systems: these are qualitatively new catalytic technologies, including those based on catalytic microreactors.

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