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Current Trends in the Application of Monolithic Catalysts in Environmentally Friendly Technologies for Autonomous Heat Supply and Post Combustion Emission Control: A Mini Review

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Abstract

Several approaches aimed at the development of catalytic packages based on ceramic and metal monolithic materials are reported and discussed in this paper. These approaches are intended for heat-generating devices and exhaust gas purification systems where high efficiency and environmental reliability are required with a wide range of concentrations and chemical natures of ecotoxics, and where the inlet flows are characterized by high volumetric velocities. In particular, the development of an efficient solution-combustion synthesis (SCS) providing more stable and active catalytic coatings on monolith walls is discussed. For effective and environmentally clean local heat supply, a technological solution is proposed involving two-stage combustion of gaseous fuel. At the first stage, the fuel is burned in a flame under oxygen-deficient conditions to suppress the formation of toxic nitrogen oxides. At the second stage, the combustion products are diluted with air, and the resulting hot gas is purified from combustible pollutants on honeycomb-structured ceramic catalysts. Such devices are used for heating greenhouses, cowsheds, residential buildings under construction, and various industrial facilities. Industrial monoliths have a limited diameter, typically not exceeding 300÷500 mm. This is the main limiting factor in the application of catalytic technologies based on monolith materials for the purification of exhaust gases from high-power internal combustion engines. For the purification of the exhaust gas from a stationary diesel power station of 2,000 kW power with a nominal exhaust flow rate of 30,000 Nm³/h, monolithic catalysts and a modular catalytic reactor comprising several monoliths of a large size have been developed.

1. Introduction: PRO et CONTRA for monolithic catalysts application

The development of technologies based on honeycomb-structured catalyst (HSC) was spurred by their widespread use in automotive exhaust gas converters in the United States in the 1970s [1, 2]. Today, the use of honeycomb catalysts and their

alternative – structured high-permeability catalysts, is not limited to automotive transport (three-way catalyst for gasoline cars and diesel fuel oxidation catalyst for diesel trucks) but is also addressed to environmental issues related to off-gas purification from stationary and mobile sources [3–5] and alternative eco-friendly heat-generating devices [6, 7] (Fig. 1). Examples of environmentally friendly applications of HSC include selective catalytic reduction of nitrogen oxides, such as in power plants, marine engines, and waste incineration plants. The energy

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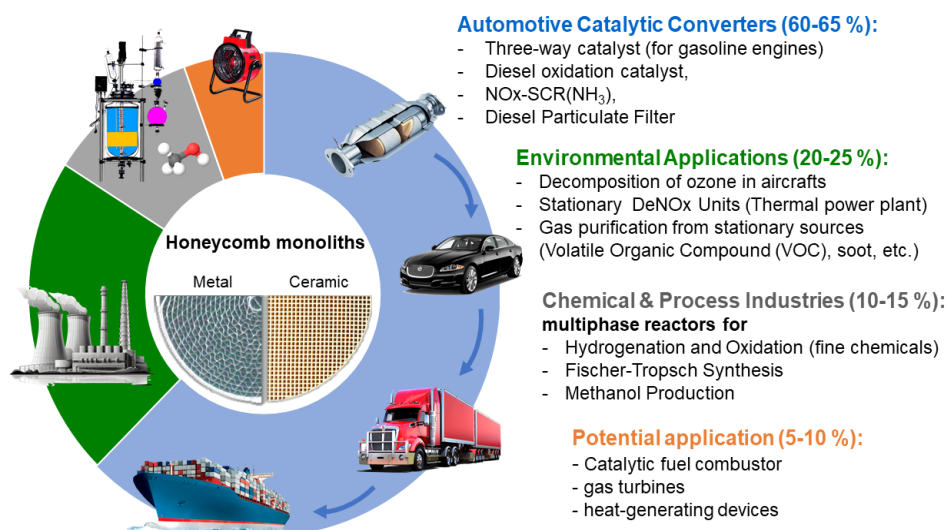


Fig. 1. Areas of practical (industrial) application of monolithic catalysts.

niche of honeycomb catalyst application also includes catalytic heat generators operating on the principle of two-stage combustion of gaseous fuels for local heat supply, for example, for greenhouses, various industrial and agricultural facilities.

However, despite numerous efforts of R&D groups in optimization of the obvious advantages of honeycomb-structured catalyst in various chemical reactions (reactive chemical distillation of methyl acetate to acetic acid and methanol [8], water distillation to enhance the esterification of alcohol with carboxylic acid [9], CO and H₂ synthesis [10–12], etc.), only a few examples of large-scale industrial application of monolithic catalysts are known [3, 4]. The most successful example is a multiphase process for the production of hydrogen peroxide using the anthraquinone process [13]. According to publicly available sources, over 100,000 m³ of monolithic supports and catalysts are produced worldwide annually to meet demand in these and other related applications. Manufacturers of honeycomb monoliths for various applications [open internet sources <https://www.accio.com/plp/honeycomb-monolith>; <https://www.corning.com/worldwide/en/the-progress-report/crystal-clear/global-warming-stands-face-to-face-with-honeycomb-technology>, etc.] predict that the global honeycomb catalyst market will grow at a CAGR of 6.8% from 2023 to 2030, with 68% of applications still in automotive catalytic converters because of tightening emission standards.

The expansion of the application range of monolithic catalysts has become possible because of a number of their physical and physicochemical properties, in particular high mechanical strength, high resistance to elevated temperatures and ther-

mal shock, low coefficient of thermal expansion, low gas-dynamic resistance to gas flow, and others [2]. The main advantages and disadvantages of practical application of honeycomb monoliths and catalysts based on them are summarized in Table 1.

To date, scientific approaches have been developed for preparing monolith-type supports with high thermal stability [2, 3], for the extrusion of ceramic monoliths with a honeycomb structure [2, 3, 7, 14–16], and for the application of catalytically active compositions to the surface of honeycomb supports [1, 2, 4, 5, 6, 15–22]. Modern honeycomb materials are produced from cordierite, various alumina modifications, silicon oxide, silicon nitride and carbide, and other inert refractory oxides, although cordierite is still the most widely used honeycomb monolith material because of its demand in automotive catalytic converters. Cordierite honeycomb supports are available with varying channel densities (from 200 to 1200 channels per square inch (cps) [2, 4]), which allows for targeted variation of the geometric surface area to volume ratio of the monolith (from 1850 to 3450 m²/m³ [2, 3]) and adjustment of catalyst performance by increasing the amount of applied catalytically active coating. The pressure drop for monoliths, even with high cell density, compared to fixed bed of catalyst granules is quite low – typically 10 times lower [4].

The main disadvantage of cordierite monoliths is their low specific surface area. To increase this, the monolith surface is coated with an inert material with a high specific surface area, such as Al₂O₃ [2, 4, 16], or a catalytically active coating. Commonly used application methods, known as washcoating, include impregnation of the monoliths with a suspension

[3, 23] or colloidal solution [3, 24], sol-gel application [25], followed by thermal treatment to convert the precursors into Al_2O_3 and SiO_2 . Catalytically active metals, including Pt-group metals and transition metal oxides, are applied to an inert coating using available methods (incipient wetness impregnation, sorption, ion-exchange, deposition-precipitation, etc. [3]). The catalytic efficiency of a washcoated catalyst

is limited by the thickness of the washcoating with the applied catalytically active metals, which cannot be increased beyond 40 μm [3] because of reduced adhesion strength. Low adhesion strength reduces catalyst service life due to catalytic coating wear and reduced catalytic efficiency and generally complicates the use of washcoated monolithic catalysts in devices with high volumetric gas flow rates.

Table 1. The advantages and disadvantages of honeycomb monoliths and monolithic catalysts

	Advantages	Disadvantages
Honeycomb monolith support (cordierite and other ceramic materials)	• high geometric external surface, variable through density (200–1200 cpsi) and shape of channels • considerable resistance to thermal shocks (up to 1350 °C) • chemically stable in oxidizing and reducing environments • high melting temperatures (1465 °C) • high mechanical strength • structural durability • short diffusion lengths • good mass transfer • low pressure drop • uniform flow distribution within the matrix • good washcoat adherence • ease of scale-up • cost-effective production	• low specific surface area, about 0.3-1 m^2/g • large pores • there is no catalytic activity • limited diameter, typically not exceeding 300÷500 mm
Honeycomb monolithic catalyst (bulk)	• excellent catalytic activity at significant GHSV ($> 50000 \text{ h}^{-1}$) • high specific surface area, more than 100 m^2/g • wide range of cell densities • good mechanical strength • high mass transfer rates • easy catalyst separation • extended service life (at least twice as long as when using granulated catalysts)	• possibility of slip of toxic components without neutralization due to the peculiarities of hydrodynamics in the channels • insufficiently high thermal resistance (depending on the material) • increased deactivation by catalytic poisons due to the lower mass of the active component • loss of catalytic activity due to transformation and recrystallization to materials with low surface area
Honeycomb monolith with washcoated catalyst	• moderate catalytic activity • high specific surface area of washcoat, more than 200 m^2/g • multilevel (micro- /macro-) porous structures • relatively good thermal stability (up to 1000 °C) • high mass transfer rates • lower volume of catalytic load (approximately 25% less than that of granular samples)	• limited washcoat thickness (about 15-40 μm), resulting in reduced catalytic efficiency • possibility of breakthrough of toxic components without neutralization due to the lower mass of the active component per unit volume and the peculiarities of hydrodynamics in the channels • increased deactivation by catalytic poisons due to the lower mass of the active component • insufficiently high thermal resistance (not more than 1000 °C) due to transformation and recrystallization of washcoat to materials with low specific surface area • detachment of washcoat due to hot exhaust gases of high velocity and mechanical vibrations

Industrial monoliths have a limited diameter, typically not exceeding 300÷500 mm. This is the main limiting factor in the application of catalytic technologies based on monolith materials for the purification of exhaust gases from high-power internal combustion engines (e.g., marine engines, etc.) and heat generators.

Below, we discuss several approaches to the development of catalytic packages based on ceramic and metal monolithic materials proposed by researchers from scientific schools led by Z.R. Ismagilov (Boreskov Institute of Catalysis, Novosibirsk, Russia), Z.A. Mansurov, and K.D. Dosumov (Almaty, Kazakhstan). These approaches are intended for heat-generating devices and exhaust gas purification systems where high efficiency and environmental reliability are required with a wide range of concentrations and chemical natures of eco toxicants, and where the inlet flows are characterized by high volumetric velocities.

2. Solution combustion synthesis for active component deposition on honeycomb monoliths

An analysis of the scientific and patent literature reveals that developments in monolithic catalysts for air heaters since 2010 have been relatively few and sporadic. Similar honeycomb catalysts for heat-generating units include commercially available monolithic honeycomb catalysts produced by Johnson Matthey, BASF Catalyst LLC, and Catalytic Products International, whose active components are platinum-group metals, primarily Pd. These catalysts are used for the combustion of gaseous hydrocarbon fuels and the purification of industrial emissions from various industries. However, catalysts based on noble metals lose activity at temperatures above 700 °C, which is unacceptable from the standpoint of efficiency and long-term operation of catalytic heat generators and gas emission purification devices.

Ceramic honeycomb supports usually have a surface area not exceeding 10 m²/g. This surface area is insufficient for the formation of nanosized particles of the active component and for ensuring high catalytic activity. The use of washcoating for catalyst preparation significantly complicates the catalyst preparation technology and is cost-intensive due to secondary coating loss during operation. To achieve high dispersion of the active component with a narrow particle size distribution on the surface of the sintered ceramic material, unconventional synthesis methods are employed. These include depositing colloidal particles of stearates and palmateates of certain transition metals (Co, Ni, Fe, Cu, etc.) in alcohol on the support surface followed by decompo-

sition of these salts [26, 27], electrolytic deposition of metals [28], synthesis under supercritical conditions [29], plasma chemistry [29–32], and diverse variations of self-propagating high-temperature and low-temperature synthesis (SHS and SLS, respectively). Nanometer-scale catalytically active particles are preferred because of so-called "size effects," which provide the nanoscale object with unusual chemical and physical properties, including catalytic activity. Some of the above-mentioned methods for dispersing oxide and metallic nanoparticles of transition and noble metals are technologically labor-intensive or impracticable when applied to bulk ceramic monoliths.

From a practical standpoint, existing variations of self-propagating synthesis hold promise because of their simplicity and low cost. Self-propagating high-temperature synthesis, first proposed by Merzhanov [33, 34], is characterized by low energy consumption, short synthesis times, and the possibility of single-step conversion of precursors to final products using the chemical energy of an exothermic reaction [33–35], which has determined its widespread use for the production of various materials [36–39]. The main limitations of SHS for the preparation of supported catalysts with catalytically active particles are associated with the development of extremely high temperatures and the difficulties of controlling the extreme process conditions. Because of the high temperatures of the SHS wave (>2000 K), it is practically impossible to obtain nanosized particles, and the final product of the synthesis is often massive materials with a low specific surface area.

A low-temperature modification of SHS, the so-called solution combustion synthesis (SCS) was proposed by Kingsley and Patil [40, 41]. The essence of SCS (Fig. 2) lies in the occurrence of an exothermic oxidation-reduction reaction between the metal nitrate precursor of the active component and a suitable organic fuel (urea, carbohydrazide, maleic acid hydrazide, etc.), the nitrate anion acts as an oxidizing agent, and the organic fuel is a reducing agent. A saturated solution is preliminarily prepared from both reagents, which ensures mixing of the components at the molecular level and homogeneity of the composition of bi- and multi-component particles of the active component [42]. After initiation of nitrate decomposition (by heating to 150÷200 °C), a rapid reaction is observed because of the propagation of a thermal wave. Thermal wave temperatures of approximately 900÷1000 °C are usually sufficient to form nanocrystalline oxide materials [43–48]. The large amount of gas released limits both the strong temperature increase and the size of the active com-

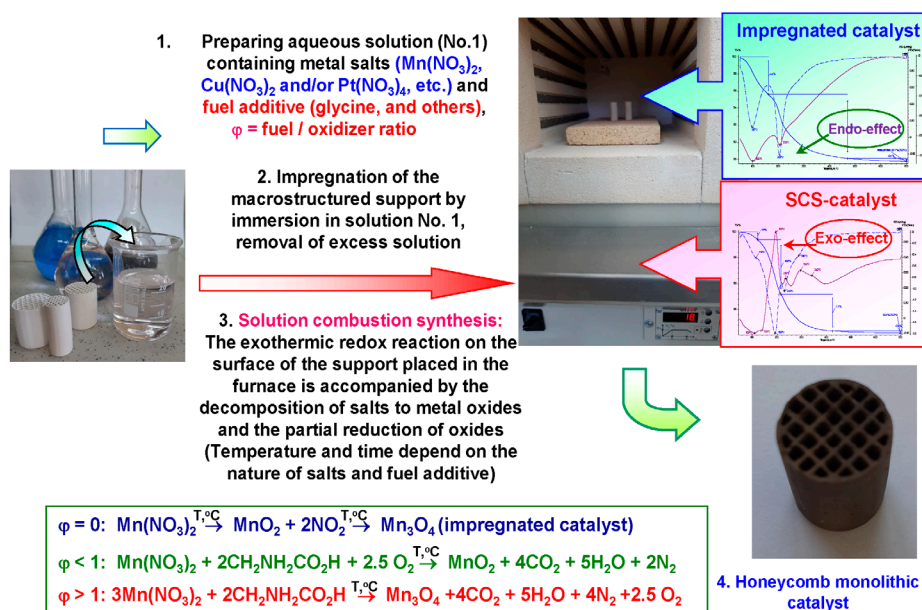


Fig. 2. Main stages of SCS for the preparation of catalysts on macrostructured supports.

ponent particles [41, 42]. This results in the formation of nanometer-sized particles with a high specific surface area [42, 43] and strong interaction with the support surface. These active component particles form the catalyst nanostructure (Fig. 3). It should be especially noted that the SCS method is applicable for depositing the active component onto macrostructured carriers of various geometric shapes, such as honeycomb monoliths, glass fabrics, highly porous reticulated foam materials, fiberglass-based fibrous filters, etc. (Fig. 3). All the listed carrier macrostructures have the potential for solving environmental problems in general and those of local heat supply [7, 21] in particular. In catalysts based on them, the carrier ensures mechanical strength, low gas-dynamic resistance, gas flow, and filtration. The catalytically active component ensures the efficiency and selectivity of the conversion of chemical substrates. There are examples in the literature in which the self-propagating synthesis method is used to prepare catalysts on monolith-shaped supports [46–48], however, these studies are rather informative in nature, demonstrating the potential of the method.

Systematic studies on the use of SCS for the preparation of macrostructured catalysts have been published by Mansurov, Ismagilov, and Dosumov [6, 12, 49–53]. The main approaches we used to improve the SCS method comprise reducing the content and increasing the dispersion of active components deposited on the surface of sintered ceramics of various geometries (honeycomb monoliths, highly porous reticulated foam ceramics, glass fabric, fibrous filters, granules, etc.). Besides dispersion, the uniformity of active component deposition, its phase composition,

the interaction of active component particles with the support, and the resistance of the particles to sintering were monitored. These factors, together with catalytic activity, determine the performance characteristics of the catalysts.

A series of studies on the preparation of oxide catalysts on glass fabrics (a type of non-porous structured support), in which active sites were formed using the SCS method, was performed [12, 49, 50]. It was found that this preparation method ensures a uniform distribution of active components in the near-surface layer of the fiberglass fabric as individual nanoparticles measuring 10–20 nm, tightly bound to the carrier surface. HRTEM, H_2 -TPR, and UV-Vis DR methods were used to determine the chemical and structural composition, which could not be determined using X-ray diffraction because of the nanoscale size of these particles (Fig. 3, "Nanostructure" inset). It was found that, depending on the molar ratio of Ni and Co oxides, the active component is formed as spinels of Co_3O_4 and $(\text{Co}, \text{Ni})\text{Co}_2\text{O}_4$ or NiO . Analysis of the diffuse reflectance UV-Vis spectra suggested the formation of defective spinel phases $[\text{Co}^{2+}, \text{Ni}^{2+}](\text{Co}^{3+})_2\text{O}_4$ and $[\text{Co}^{2+}](\text{Al}^{3+}, \text{Co}^{3+})_2\text{O}_4$ on the glass fabric surface and confirmed the H_2 -TPR data on the strong interaction of $\text{Ni}^{2+}_{\text{Oh}}$ and $\text{Co}^{2+}_{\text{Td}}$ cations with the support. The observed high-temperature shift of the hydrogen consumption maximum indicated the interaction of the active components with the support up to the formation of new phases. Thus, active spinel phases based on $[\text{Co}^{2+}](\text{Co}^{3+})_2\text{O}_4$ were formed at moderate temperatures, about 400–450 °C versus 700 °C using the incipient wetness impregnation method.

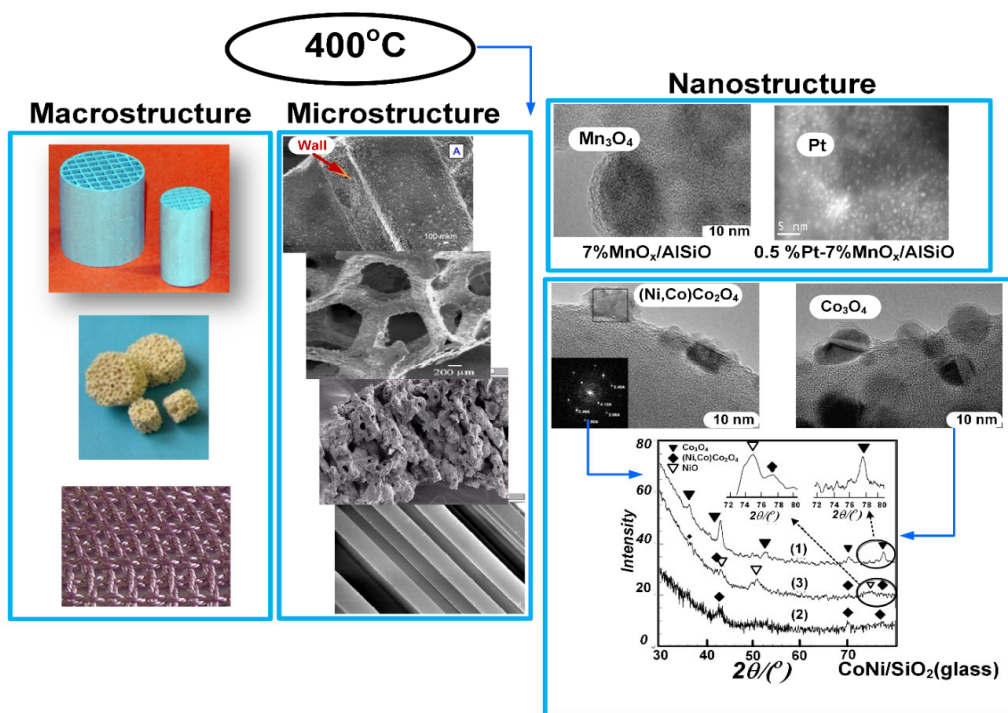


Fig. 3. Macro-, micro- and nanostructures of catalysts obtained using SCS.

Catalytic systems with a spinel structure demonstrated high activity and stable operation in the oxidation reactions of hydrocarbons, CO [49] and carbon dioxide reforming of methane [12, 50]. It was shown that spinel nanoparticles of the active component prevent carbon formation and ensure high activity in the process of carbon dioxide reforming of methane to synthesis gas with a ratio of $H_2/CO = 1/1$ [50]. Even at a low concentration of the catalytically active component (near 0.3–1.5 wt.% on CoO [49]), the catalyst ensured high activity in the oxidation of methane, butane and CO, which increased linearly with increasing CoO content up to 1.2 wt.% [49] (Fig. 4) and was not subject to deactivation in a long-term experiment [50].

The SCS method was tested for the preparation of monolithic catalysts for various purposes, in particular for the deep oxidation of carbon monoxide and C_1 – C_4 hydrocarbons. The honeycomb monoliths used were based on sintered Al_2O_3 – SiO_2 ceramics and had a low specific surface area (not more than $13.5 \text{ m}^2/\text{g}$) and pore volume (not more than $0.25 \text{ cm}^3/\text{g}$). Its porous structure included meso- and macropores with a predominance of pores up to 500 nm in size (86%). The SCS method made it possible to apply highly dispersed nanoparticles of the active component to this ceramics, consisting of individual (Mn, Fe, Ni, Co, Cu), mixed (Fe–Mn, Ni–Mn, Co–Mn, Cu–Mn) and modified (Mn–Pt, Mn–Pd, Mn–Pd–Pt, Mn–La, Mn–Ce, Co–La, Co–Ce) oxides [6, 51–53]. The structural, textural, morphological, and oxidation-reduction properties of the

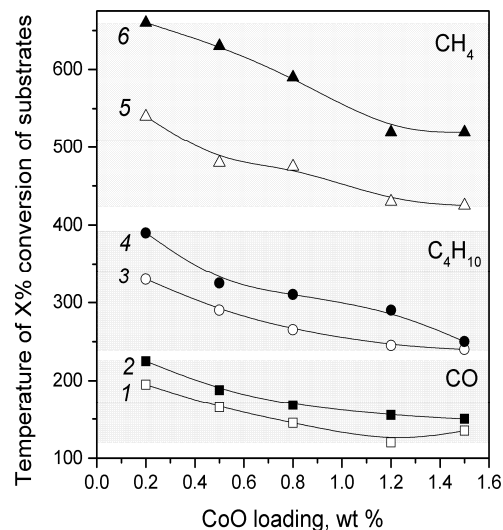


Fig. 4. Temperature of 50% (1, 3, 5) and 90% (2, 4, 6) conversion of CO (1, 2), C_4H_{10} (3, 4) and CH_4 (5, 6) on Co/ SiO_2 (glass fabrics) as function of CoO loading. Substrate oxidation condition: 1 vol.% CO, CH_4 , or C_4H_{10} in air; gas hourly space velocity, 1000 h^{-1} . The data are adopted from [49, 50].

catalysts were studied using a set of physicochemical methods. The features of the formation of the nanostructure of the active components on the sintered surface of the monolithic support in single-component and two-component catalysts were revealed, depending on the chemical composition of the catalyst. Using X-ray diffraction and HRTEM methods, it was established that the active component parti-

cles in catalysts based on individual oxides of Mn, Fe, Co, Ni, and Cu are well-crystallized oxide phases with a particle size of 100–150 nm and varying degrees of interaction with the support [50]. According to HRTEM, H_2 TPR, UV-Vis DRS, and differential dissolution data, the active component particles of Fe-Mn, Ni-Mn, Co-Mn, and Cu-Mn catalysts are represented by a mixture of simple oxide phases, weakly crystallized, highly defective complex oxide phases with numerous interparticle boundaries, and a phase of interaction of transition metal cations with the support materials [6, 51]. Depending on the catalyst composition, the particle size of the active component varies from 20 to 100 nm, whereas, the introduction of transition metal oxides into the support matrix by the method of incipient wetness impregnation ensured the formation of coarse particles larger than 200 nm, despite their uniform distribution over the geometric surface of the support.

It was found that the addition of La and Ce oxides allows the dispersion of MnO_x - and CoO_x -containing active component particles to 10–25 nm, which is 2–3 times less than in impregnated samples of similar composition [51, 52]. A similar trend is observed when MnO_x particles are modified with platinum. The MnO_x nanoparticles in the ex-SCS sample are 2–3 times smaller than in the impregnated sample, 10–15 nm versus 20–50 nm (Fig. 5). The localization depth of catalytically active oxides in SCS catalysts is lower than in impregnated catalysts. The morphology

of particles formed under SCS and incipient wetness impregnation conditions is different. In SCS samples, oxide particles have a broad base immersed in the subsurface layers of the support, while in impregnated catalysts, oxide particles interact weakly with the support (Fig. 5, “Morphology” inset). HRTEM, UV-Vis DR, and H_2 -TPR analysis showed that the use of SCS resulted in the stabilization of a higher amount of low-oxidation transition metal oxides (Mn_3O_4/MnO (Fig. 5, “RedOx and catalytic properties” inset), Co_3O_4/CoO), with their proportion in the catalyst increasing with the amount of fuel additive (glycine), and in an increase in defects when modified with La or Ce. UV-Vis DR analysis showed that some high-oxidation transition metal oxide (Mn_2O_3 , MnO_2) was present on the surface [51, 52]. H_2 -TPR experiments showed that the proportion of Mn_3O_4/Co_3O_4 was higher in Ce oxide-modified $Mn(SCS)/AlSi$ catalysts compared to La-oxide-modified samples and the unmodified sample. It was shown that under SCS conditions, the active components are formed as highly dispersed particles of metallic Pt and manganese oxides Mn_3O_4 , located in the surface layers of the support (Fig. 5, “Size of active component particles” inset). In contrast, during thermal treatment of the impregnated catalyst (for example, MnO_x -Pt, Fig. 5), the resulting manganese oxides are enriched in Mn(IV) cations and are predominantly localized within the bulk of the support (Fig. 5, differential dissolution data), forming an interaction phase with it (HRTEM, Fig. 5).

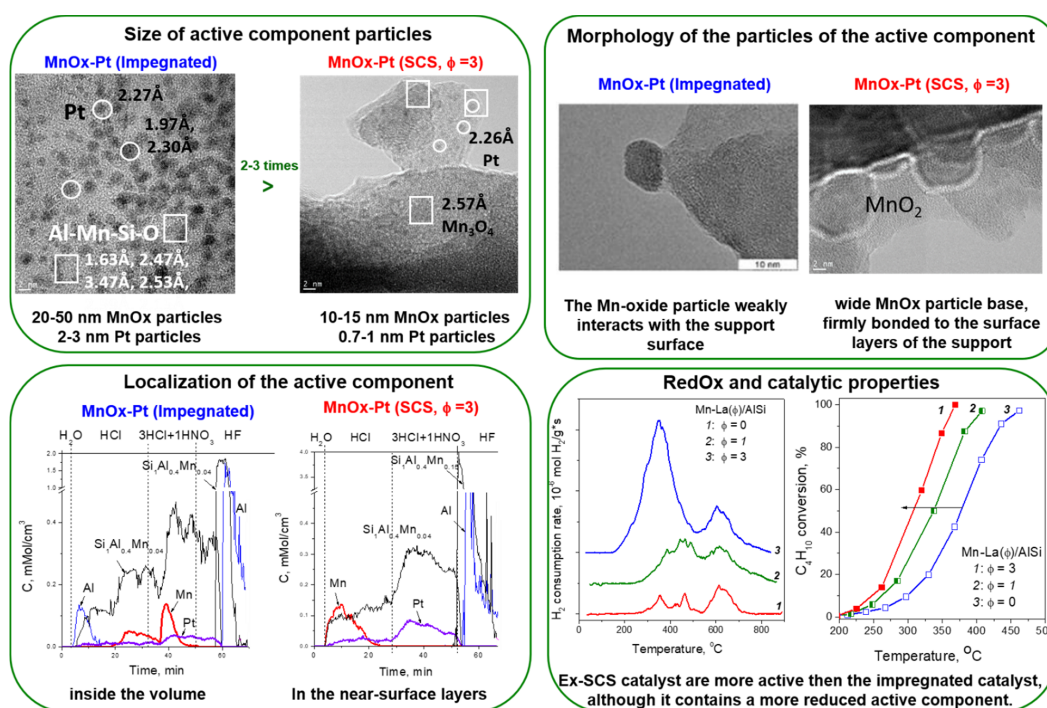


Fig. 5. Interdependencies “synthesis conditions” – “structure” – “properties” for catalysts obtained by incipient wetness impregnation and SCS.

Relationships have been established between the structure of the active component in one- and two-component catalysts and the catalytic activity in the oxidation reaction of methane, butane, and carbon monoxide [51–53]. It has been shown that the synergistic effect in two-component catalysts is because of the formation of highly defective complex oxides in the surface layers of the support: $\text{Cu}_2\text{Mn}_3\text{O}_8$, NiMnO_3 , and/or solid solutions of Ni(II) cations in MnO_2 , Fe(II,III) cations in Mn_2O_3 , enriched with Mn^{4+} cations, as well as the stabilization of manganese oxides in valence (IV,III) [6, 50].

It was shown that in the case of catalysts containing the same active component Mn-La, Mn-Ce, Co-La or Co-Ce the catalysts prepared by the SCS method were more active in the reaction of butane oxidation than those prepared by impregnation (Fig. 5, “RedOx and catalytic properties” inset). Generally, their activity grows with an increase in the content of fuel additive from $\varphi = 0.7$ to $\varphi = 3.5$ (Fig. 6). The best stability (at 450 °C) was demonstrated by catalysts prepared by SCS in an excess of glycine. They provided the butane conversion at a level of 85% after 100 hours of testing [52]. Correlations composition-structure-localization of active oxides and the catalytic activity of SCS catalysts were elucidated. The high activity of SCS catalysts in butane oxidation is provided by the enrichment of the subsurface layers of the support by

manganese/cobalt oxides with low oxidation states ($\text{Mn}_3\text{O}_4/\text{Co}_3\text{O}_4$), high defectivity of oxide structures, and developed pore structure [51, 52]. The high activity of SCS catalysts of the Pt- Mn_3O_4 composition in butane and methane oxidation reactions is determined mainly by the presence of reduced forms of manganese and the availability of active components for reactants in the near-surface layers of the support [53].

Catalytic systems promising for practical application in autonomous catalytic heaters were selected. Among the selected systems are those based on oxides of cobalt and manganese, including Co and Mn catalytic systems modified by oxides of copper, lanthanum, or cerium. Testing of an experimental batch of the full-size catalyst Mn-La/AlSi was conducted in a test bench comprising a direct gas-fired air heater of the suspended series “Fermat T-80C”. It was found that the continuous operation of the air heater did not lead to an increase in the pollutants in the work area above the maximum allowable limits.

3. Catalytic heat generators for local energy supply

Among autonomous (local) heating devices capable of significant cost reduction in the transportation, distribution, and consumption of thermal energy, catalytic heat generators are recognized as the most efficient and environmentally friendly.

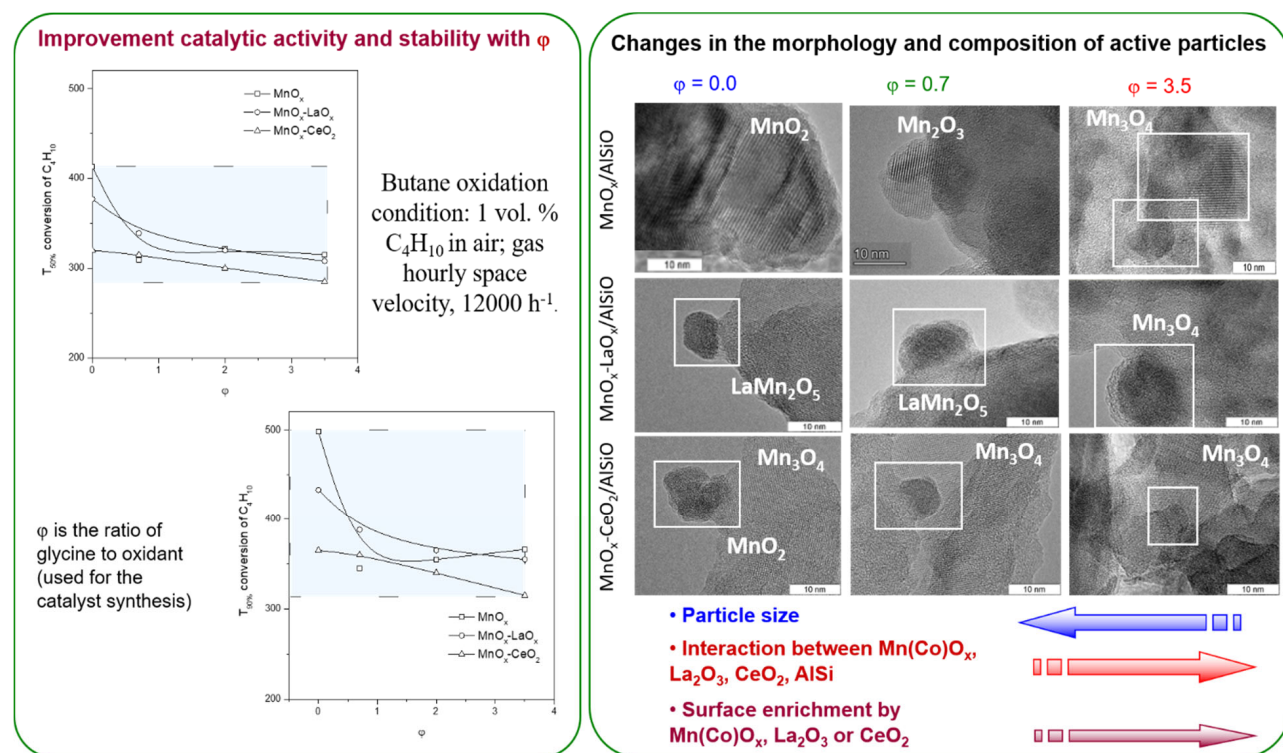


Fig. 6. Effect of the ratio of glycine to oxidant (used for the catalyst synthesis) on interdependencies “ φ ” – “structure” – “properties” for MnO_x , $\text{MnO}_x\text{-LaO}_x$ and $\text{MnO}_x\text{-CeO}_2$ catalysts obtained by incipient wetness impregnation and SCS.

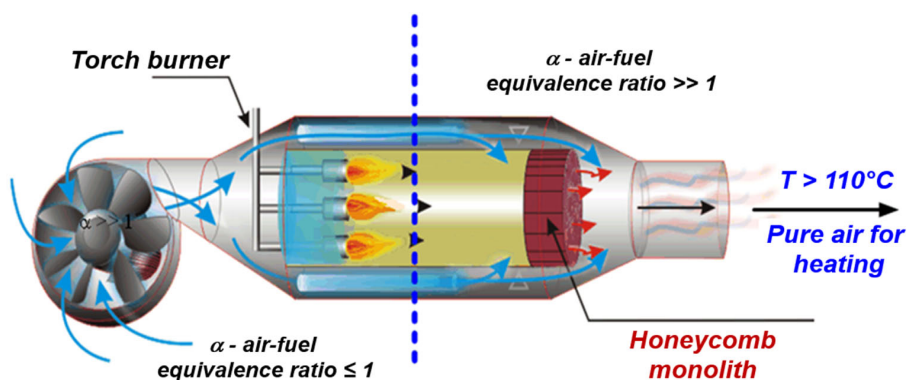


Fig. 7. Technical concept of a two-stage catalytic heater.

The main trends in the development of autonomous heat sources are the improvement of traditional combustion devices: optimization of burner operation, development of microprocessor-based fuel/air ratio control devices, and optimization of the overall engineering design of heat generators. Despite significant efforts and thorough research in this area, there are a number of fundamental limitations that prevent the creation of devices with a thermal capacity of over 100 kW, providing DIRECT HEATING of premises and drying of agricultural products. The primary shortcoming is the intensive formation of nitrogen oxides at flame temperatures $> 1200^{\circ}\text{C}$, typical for all burner devices.

The proposed technological solution involves two-stage combustion of gaseous fuel (Fig. 7). In the first stage, the fuel is combusted in a burner flame under oxygen-deficient conditions to suppress the formation of toxic nitrogen oxides. Combustion of gas fuel at an air-fuel equivalence ratio $\alpha \leq 1$ reduces nitrogen oxide concentrations to an acceptable level of 1–5 ppm (Fig. 8, red line), eliminating the need for expensive DeNO_x combustion product purification systems. In the second stage, the combustion products are diluted with air ($\alpha \gg 1$), and the resulting heated gas, at a temperature of 500–650 $^{\circ}\text{C}$ is fed to a catalytic converter, a composite element made of honeycomb-structured ceramic catalysts. Here, a catalytic converter burns the incomplete combustion products to non-toxic compounds (CO_2 and water). As a result, the content of toxic compounds (mainly CO and hydrocarbons) is reduced (Fig. 8, blue and black lines) to at least the level of sanitary standards for work species. After passing through the converter, the purified gas is further diluted with air, and the resulting gas, at a temperature of 100–120 $^{\circ}\text{C}$, is used directly as a heat transfer fluid for space heating.

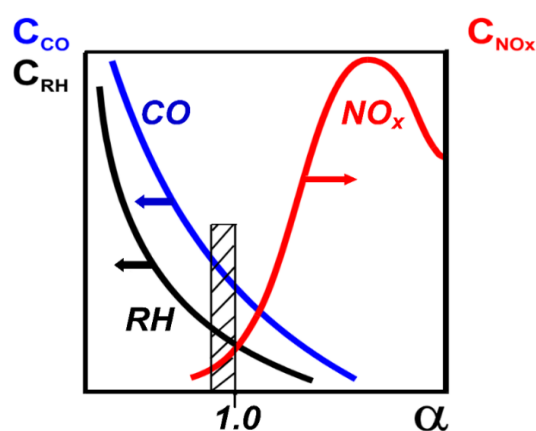


Fig. 8. Formation of NO_x, carbon monoxide (CO) and “underburning” of initial hydrocarbon (RH) upon variation of air-fuel equivalence ratio, $\alpha = 1$ corresponds to stoichiometry.

In practice, existing commercially produced heat guns were equipped with a special catalytic after-treatment unit, consisting of a metal shell and a cassette containing honeycomb catalyst monoliths with a hexagonal cross-section. The assembled cassette was optimally adapted to the dimensions (diameter) of the heat gun, and its location ensured a temperature of 700–750 $^{\circ}\text{C}$ at the frontal surface of the catalyst. The use of catalysts enables the production of a heat transfer fluid with toxic component levels significantly lower than maximum permissible concentrations (MPCs), suitable for direct heating of residential and work spaces and for drying agricultural products. This completely eliminates the need for secondary heat transfer fluids.

A key feature in the design of developed devices is the use of catalysts, the specific application of which determines necessary characteristics such as high catalytic activity, resistance to catalytic poisons, high efficiency in removal of pollutants, and, above

all, low pressure drop. Honeycomb catalysts fully satisfy these requirements. Analogues of the developed honeycomb catalysts are industrially produced monolithic catalysts from Johnson Matthey, whose active components are platinum group metals (Pt, Pd). Such catalysts are used for the combustion of gaseous fuels and the purification of industrial emissions from various industries. However, catalysts based on noble metals lose activity at temperatures above 700 °C, which is unacceptable from the standpoint of the efficiency and long-term operation of catalytic heat generators and emission purification devices. An alternative to catalysts based on noble metals is oxide systems, which retain high activity at high temperatures [5, 54].

Examples of thermally stable granulated oxide catalysts are those based on manganese oxides and aluminum oxide modified with La_2O_3 , CeO_2 , and MgO additives, developed at the Boreskov Institute of Catalysis SB RAS [55, 56]. The stabilization of manganese oxide in alumina structure using the thermodispersion effect increases the activity of the granular catalyst in the methane oxidation [7, 55]. These

catalysts can operate at elevated reaction medium temperatures of up to 1000–1100 °C [54]. However, granular catalysts have inherent drawbacks, primarily high gas-dynamic resistance, which limits their use in the treatment of gas streams with a high flow rate. Based on the knowledge about the catalytic behavior of manganese-alumina catalysts in the deep oxidation reactions of methane and other hydrocarbons [7, 54–56], monolithic catalysts containing manganese oxides as an active component have been developed [51–53, 57, 58]. Comparative characteristics of the developed catalysts [57, 58] and their analogues [59] are presented in Table 2. It can be seen that the monolithic catalyst with deposited Mn_3O_4 nanoparticles (no more than 30 nm in size) [57] is not inferior in catalytic efficiency to the honeycomb catalyst with a Pt,Pd-active component. The advantage of a MnO_x -containing catalyst is the high allowable overheating temperature, 1100 °C versus 725 °C for its industrial counterpart.

Relative characteristics of the developed catalytic heat-generating devices (CHGDs) and industrially used heat “guns” [60, 61] are shown in Table 3.

Table 2. Comparative technical characteristics of honeycomb structured catalysts

Parameter	Developed catalysts	Fuel combustion catalyst (Honeycat Johnson Matthey, UK) [59]
Active component	Transition metal oxides (manganese, copper, iron)	Platinum group metals (platinum, palladium) PGM content $\geq 2\%$ by weight
Nominal operating temperature, °C	400–780	400–650
Permissible overheating temperature, °C	1100	725
Pressure drop at a space velocity (GHSV) of 20,000 h^{-1} and a catalyst length of 1 m, kPa	0.98–2.9	0.98–2.9
Particle size of the active component in the honeycomb structured catalyst, nm	not higher 30	higher 100
Efficiency in removing toxic pollutants at $T = 650\text{ °C}$, %		
CO	> 99	> 99
Hydrocarbons ($\text{C}_1\text{--C}_4$)	> 99	> 99
Relative cost in arbitrary unit	1	5

Table 3. Comparative technical characteristics of the developed CHGDs based on honeycomb structured catalysts

Parameter	CHGD	Flare air heater Tecnoclima (Italy) [60]	STV Flare Air Heater (France) [61]
Nominal thermal power of air heaters, kW	500	500	500
Natural gas consumption, nm^3/hour	40–42.5	47.5–50.0	52–55
Metal content, (kg/kW of thermal power)	0.4	1.5	2.5
Air heater cost. Arbitrary units	1	2.3	4.4

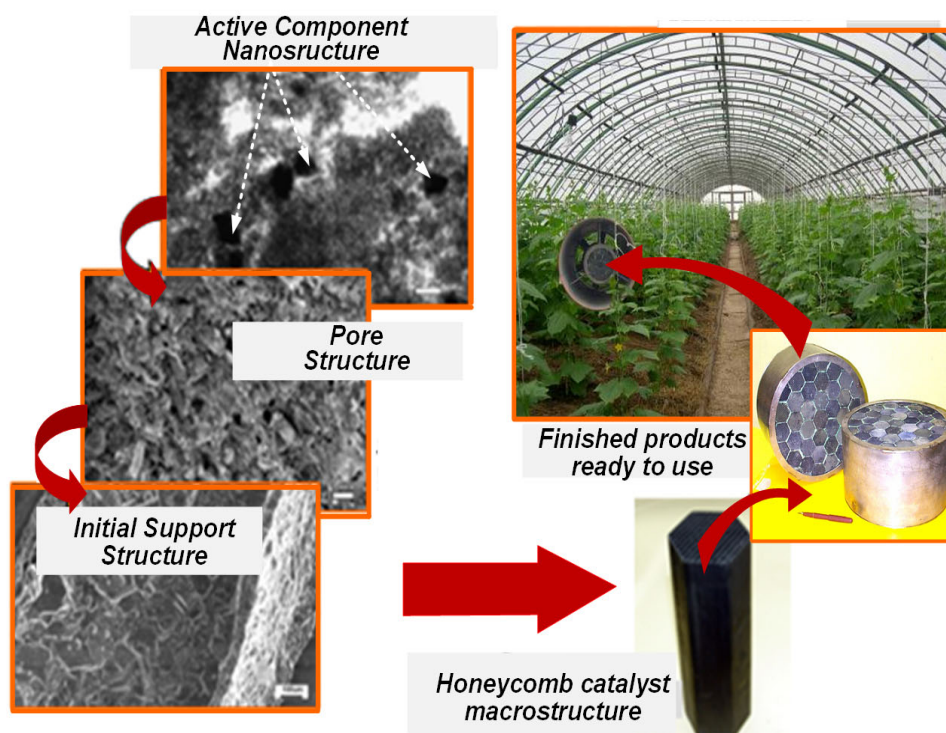


Fig. 9. The relationship between basic research and commercial product manufacturing.

Building on a substantial scientific and technical foundation, industrial prototypes of catalytic heat-generating devices (CHGDs) were developed and have been successfully operated at the Priobskoye greenhouse farm, JSC (Novosibirsk Region, Russia), since 2004. The Priobskoye farm consists of 10 greenhouses with a total volume of 22500 m³. Each greenhouse was equipped with a catalytic air heater. The key element of this device is a catalytic cassette assembled from individual monolithic catalysts of a honeycomb structure. The specifics of the preparation of catalysts (SCS) determine such characteristics as high catalytic activity, resistance to catalytic poisons, and a high degree of purification of gas emissions. The kinetic and operational properties of catalysts are largely determined by the nanoscale size of the active component particles and the porous features of the honeycomb carrier. The monolithic ceramic support provides high mechanical strength to the catalyst as a whole, thereby extending its service life.

Comparative economic indicators calculated based on the results of a long-term operation [6, 53, 60, 61] are presented in Table 4. The relative cost of greenhouse heating when using the developed CHGD is 3 times lower than the financial costs of JSC Priobskoye for heating greenhouses with thermal energy from a thermal power plant and 4 times lower if diesel-powered heat generators are used for this purpose.

Table 4. Comparative economic indicators of catalytic heating

Seasonal cost of heating greenhouses, relative costs		
CHGD	"Priobskoye" JSC budget for heating greenhouses using thermal energy from the combined heat and power plant	Diesel-fueled heat generators
1	3	4
(actual costs)		

4. Modular catalytic reactors for the purification of exhaust gases from stationary diesel power plants

The combustion products from internal combustion engines contain poisonous components: carbon monoxide, organic compounds (unburned hydrocarbons and aldehydes) and nitrogen oxides. The problem of purifying exhaust from small engines is mostly solved by developed and commercialized catalytic converters used mainly in automobiles. For the purification of exhaust from large stationary diesel engines, fabrication of monolithic catalysts of large size is required (diameter over 600 mm). Existing technologies for the production of monolithic catalysts usually provide the possibility of fabricating monoliths with diameters not exceeding 300–500 mm.

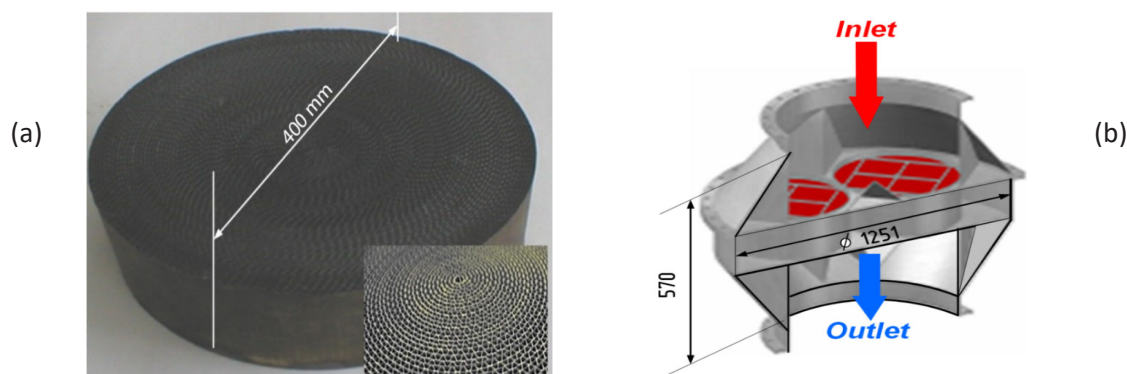


Fig. 10. The modular catalytic reactor for the purification of exhaust gases from stationary diesel power plants: (a) – single catalytic module; (b) – the sketch of modular catalytic reactor

For the purification of the exhaust gas from a stationary diesel power station of 2,000 kW power with a nominal exhaust flow rate of 30,000 Nm³/h, monolithic catalysts and a modular catalytic reactor have been developed. The drawing of the reactor is shown in Fig. 10.

The reactor contains eight cylindrical catalytic monoliths. The monoliths are installed in common cylindrical casing in consecutive pairs symmetrical with respect to the reactor cross-section. For the provision of a uniform flow of the gas through the monoliths, five gas spreaders in the form of pyramids are installed: one at the reactor center and four at the reactor wall between adjacent monoliths.

For the preparation of monolithic catalysts, substrates fabricated from flat-on-corrugated fechrall alloy foil with a thickness of 0.05 mm were used. The substrate diameter is 400 mm, and the channel density is 290 cpsi (Fig. 11).

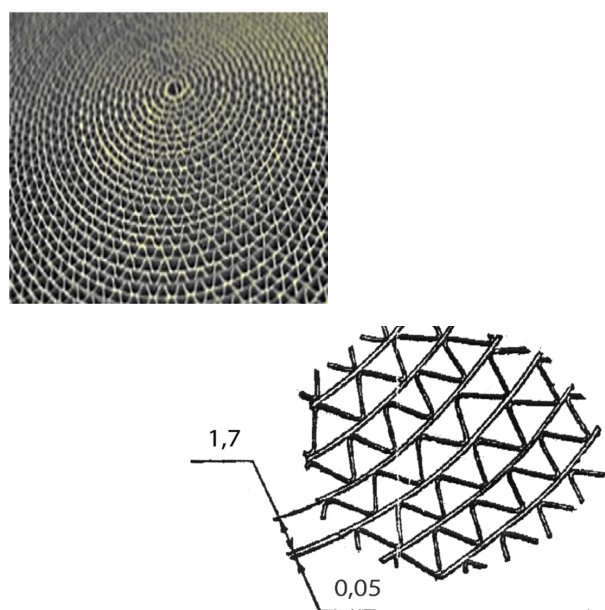


Fig. 11. The geometry of the catalyst substrate.

The monoliths were calcined at high temperature, and then the deposition of a washcoat based on alumina modified with rare earth and transition metal oxides was performed from a water suspension. The washcoated monoliths were dried and calcined at 800 °C (Fig. 12).

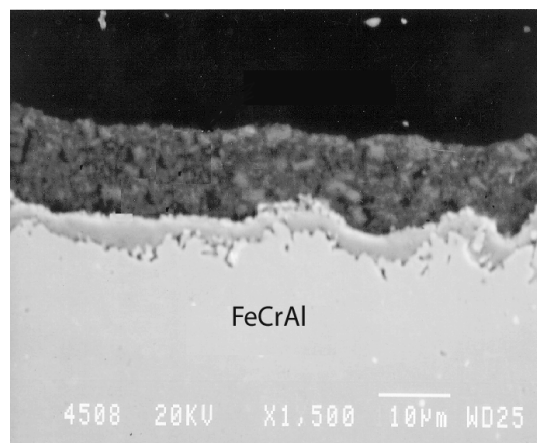


Fig. 12. Micrograph of a washcoat on the fechrall alloy.

The active component containing platinum group metals and transition metals was introduced into the washcoat by proprietary procedures. The deposition of the active component included impregnation, reduction, drying, and thermal treatment steps. The active catalyst component was deposited from aqueous solutions of precursors using incipient wetness impregnation, based on a specified platinum content of 1% by weight of the secondary support (0.1% by weight of the monolith). The impregnated monoliths were dried at 200 °C and then calcined in the presence of a reducing agent for 4 hours.

The control of the catalytic activity was performed using smaller catalyst monoliths with a volume of 5–10 cm³ prepared by the identical procedure. The conditions of the activity tests were: gas

space velocity $110,000 \text{ h}^{-1}$, the composition of the gas mixture 1800 ppm CO and 750 ppm of hydrocarbons (C_4H_{10}) in air. The typical plots of the conversion versus temperature are shown in Fig. 13. The activity of the samples was characterized by CO conversion at 400°C , and the tests showed it to be in the range of 90–93%.

At present, the developed modular catalytic reactor is fabricated and undergoes testing as part of a diesel power plant for emergency electricity supply at a subway construction company.

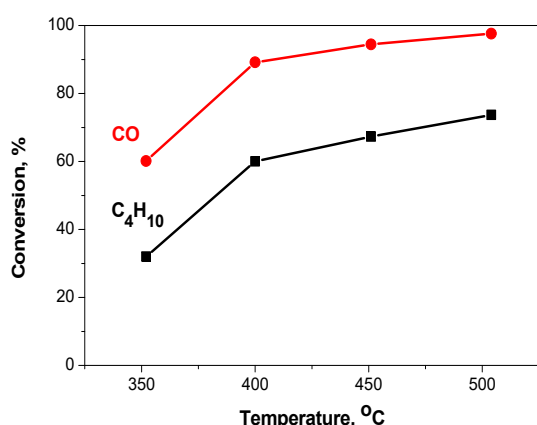


Fig. 13. Temperature dependences of CO and butane conversion over a catalyst monolith at a gas space velocity of $110,000 \text{ h}^{-1}$, the composition of the gas mixture is 1800 ppm of CO and 750 ppm of hydrocarbons (C_4H_{10}) in air.

4. Conclusion

Catalytic technologies are a recognized tool for environmental protection, and the scientifically sound selection of catalytic systems and optimal operating conditions largely ensures their operational efficiency. The common environmental and technological challenges faced by the Russian Federation and the Republic of Kazakhstan in developing such technologies provide a basis for scientific and technical cooperation, aimed primarily at developing catalysts with different chemical compositions and geometric configurations for a wide range of applications.

This review describes some examples of such successful collaboration in the development and creation of:

- catalysts produced by self-propagating synthesis for various applications;
- catalytic heat generators for local energy supply;
- modular catalytic reactor for the purification of exhaust gases from stationary diesel power plants.

The presented results are a remarkable example of how the joint fundamental research led to practical realization of the developed technologies.

CRediT authorship contribution statement

Svetlana Yashnik: Conceptualization, Methodology, Investigation, Writing – Review & Editing. **Sergey Khairulin:** Methodology, Writing – Original Draft. **Mikhail Kerzhentsev:** Investigation, Writing – Original Draft. **Anton Salnikov:** Visualization, Investigation; **Zinfer Ismagilov:** Conceptualization, Project administration. **Kusman Dosumov:** Conceptualization, Project administration, Investigation, Supervision. **Zulhair Mansurov:** Project administration, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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