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Fluidized bed catalytic combustion

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Abstract

The principles and applications of fluidized bed catalytic combustion are described. The experience and current activities of the Boreskov Institute of Catalysis in the development of fluidized bed catalytic combustion of various fuels and organic wastes are briefly reviewed. Prospects for new applications of this technique are discussed. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Catalytic combustion; Fluidized bed; Organic waste; Radioactive waste; Waste-water treatment

1. Introduction

In recent years catalytic combustion passed through the stage of multiple R & D projects into the stage of wide commercial implementation. This is caused by the fact that catalytic combustion has proved to be an effective technique to achieve clean combustion of fuels. Catalytic combustion is principally different from traditional flame combustion because organic compounds are oxidized flamelessly on the surface of solid catalysts at relatively low temperatures: 400–1000°C. This technique provides high fuel efficiency, safety and allows to drastically bring down emissions of CO, unburned hydrocarbons and nitrogen oxides [1–9]. The main applications of catalytic combustion are various domestic and industrial heat sources: radiant heaters, stoves with catalytic burners, boilers and gas turbines [6–14].

The processes of catalytic combustion can be carried out in fixed and fluidized catalyst beds. Despite the fact that fixed bed reactors are widely used in various applications they have some drawbacks limiting the intensification of the processes, namely:

1. It is difficult to extract useful heat, to provide a uniform catalyst temperature and to preclude the formation of hot spots in the fixed catalyst bed when burning stoichiometric fuel/air mixtures.
2. Fixed catalyst beds work usually with premixed fuel/air lean mixtures and the number of fuels that can be used is limited to gaseous hydrocarbons and oil distillates. It is difficult or impossible to use fixed catalyst beds for combustion of heavy oil fractions, residual oil, organic waste of complex composition and solid fuels and wastes.

In the Boreskov Institute of Catalysis the catalysts and processes for combustion of fuels and organic wastes in fluidized catalyst bed were developed [4,11–13]. The fluidized bed provides a high temperature uniformity, allows efficient heat removal and makes possible to burn fuels and organic wastes in different states of aggregation. The catalysts are a combination of metal oxides supported on spherical granules of

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special mechanically strong alumina. This method has been shown to be very effective for many applications where fixed catalyst beds cannot be used.

2. Description of technology

The process of catalytic fluidized bed combustion depends on the following four principles [4]:

1. use of catalysts for complete oxidation;
2. use of a fluidized catalyst bed;
3. stoichiometric air/fuel ratio close to 1;
4. simultaneous heat evolution and heat consumption in the same catalyst bed.

A schematic diagram showing the principle of operation of the apparatus for catalytic fluidized bed combustion – a catalytic heat generator (CHG) is given in Fig. 1. Air for fluidization and fuel combustion is supplied through a gas distributing plate. Fuel is injected into the bed through nozzles. The reaction products are removed through the upper, wider part of the apparatus. Due to the widening entrained catalyst particles separate out of the gas stream. To control bed temperature, excess heat is removed from the catalyst by heat exchangers located within the catalyst bed or, in certain cases, by endothermic processes performed inside the bed simultaneously with fuel combustion. This method of combustion provides excellent heat transfer and heat withdrawal from the bed. The design of the heat-exchange devices and of the entire reactor is relatively simple; large-sized, complicated interior and exterior heat exchangers are not required. The continuous motion of the solid particles results in heat transfer by convection, and the temperature in the bed is kept uniform. Preliminary heating of the reaction mixture to the ignition temperature of the catalyst is also not necessary, since the required temperature is quickly achieved after the gases enter the bed because of its excellent heat transfer properties.

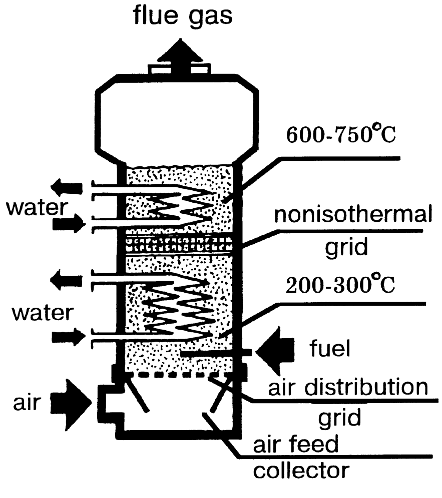

Fig. 1. Principal scheme of catalytic heat generator.

The principal advantage of CHG apparatus over catalytic fluidized bed reactors proposed earlier [15–19] is the presence of a non-isothermal grid which divides the apparatus in two zones: a lower one with the temperature of 600–750°C sufficient for complete fuel oxidation and an upper one where the temperature is determined by the conditions of heat removal and can thus be decreased to 200–300°C (Fig. 2). This design allows to decrease heat losses with exhaust gases and provides the possibility to carry out various technological processes in the bed at a controlled temperature [4,11,12].

3. Main problems encountered in the development of catalytic fluidized bed technologies

3.1. *Development of catalysts for fluidized bed combustion*

In CHG the catalyst is subjected to the following combined severe effects:

- mechanical shocks and attrition;
- high temperature;
- chemical action of stoichiometric fuel/air mixtures;
- poisoning effect of sulfur, alkaline metals and other elements in case of combustion of crude fuels and organic wastes.

Therefore the following requirements are imposed on these catalysts, namely:

- high mechanical strength;
- high thermal stability;
- conservation of sufficient activity during operation for long periods of time in media with low oxygen concentration;
- resistance to catalytic poisons.

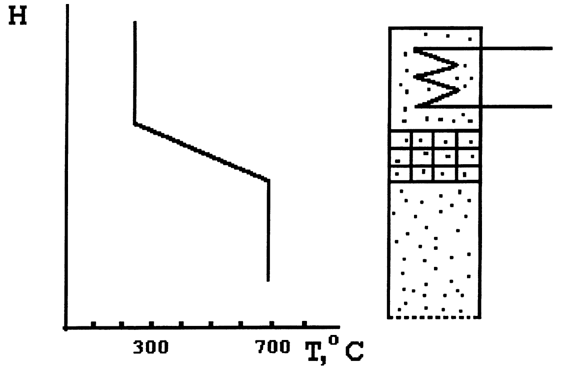

Fig. 2. Temperature profile along catalytic fluidized bed reactor with non-isothermal grid.

One of the most important problems encountered in the development of fluidized bed combustion is catalyst attrition. To solve this problem special mechanically strong (crushing strength up to 50 MPa) catalysts were developed [13]. The catalysts are prepared by supporting oxides of Fe, Cr, Mg, Cu and other metals on strong spherical alumina beads with high specific surface area. Spherical form results in less abrasion of catalyst particles in fluidized bed operation.

A study of a series of complete oxidation catalysts recommended for catalytic fuel combustion showed that in actual conditions of the process changes in their catalytic and structural–mechanical properties take place due to the action of high temperatures and reaction medium, resulting in catalyst deactivation and deterioration [4,21–24]. The principles for the selection of combustion catalysts for operation in fluidized bed, taking into account the interactions of catalyst components upon thermal treatment were formulated:

- an active component should not have a mineralizing effect on support;
- catalysts should retain high mechanical strength after thermal treatment at 700–1200°C;
- the interaction of an active component with a support should lead to granule strengthening;
- the products of the interaction of an active component with a support should have a sufficient catalytic activity for the complete oxidation of organic compounds.

On the basis of these principles and the results of a study of the complex interactions of oxide compounds of transition metals with various alumina supports a series of novel combustion catalysts with high mechanical and thermal durability, different in particular catalytic properties was developed [20,24–27]. The supports used in the catalyst preparation were spherical granules of γ-alumina of various size fractions: 0.4–1.0, 1.0–1.6, 1.4–2.0 and 2.0–3.0 mm.

A series of catalysts for CHG are produced commercially in Novosibirsk.

Further research for the development of improved catalyst and catalyst supports with increased strength and thermal durability is in progress [28–30].

3.2. *Fuel nitrogen oxides control*

A very attractive feature of catalytic combustion is the possibility to reduce drastically NO_x emissions. Sufficiently low temperature of catalytic combustion (600–750°C) in CHG allows nearly complete elimination of thermal NO_x, which are formed from molecular nitrogen at high temperature via the Zeldovich mechanism. The problem of fuel NO_x control cannot be as reliably solved for the processes of catalytic combustion, because fuel-bound nitrogen can be converted to NO_x at both high and low temperatures as was shown in many studies made with fixed beds [31–33].

Detailed kinetic studies of catalytic oxidation of nitrogen containing compounds of various types (pyridine, acetonitrile, nitromethane, dimethylformamide) were carried out in order to elucidate the regularities of fuel nitrogen oxides formation from fixed nitrogen [34–39]. The kinetic parameters of the reactions of oxidation of N-containing compounds and those of NO_x formation were measured. It has been shown that the nature of the catalyst and its qualitative and quantitative composition has the most pronounced effect on the yield of NO_x, e.g. oxide catalysts favor a much lower NO_x yield from fixed nitrogen in comparison with noble metal catalysts (Fig. 3). The parameters of the process have also a strong effect on NO_x formation: the concentration of fuel NO_x can be decreased by operation at a low excess of air and by decreasing the temperature. These results obtained in laboratory installations were confirmed by experiments on combustion of simulated and real nitrogen containing fuels in CHGs [1–3,39]. It was shown that by selection of optimum catalysts and process parameters the degree of conversion of fuel-bound nitro-

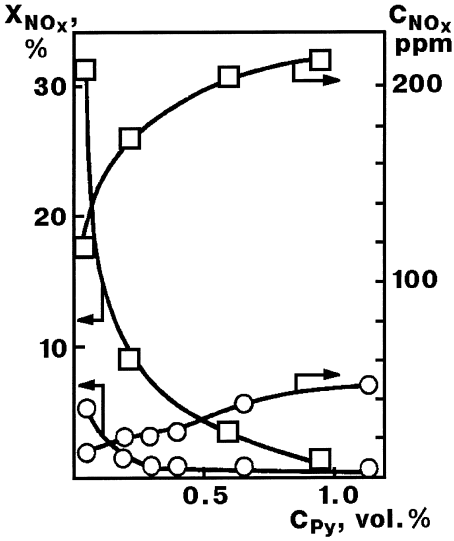

Fig. 3. Plot of NO_x concentration and conversion of fixed N to NO_x at 400°C vs. initial concentration of pyridine (vol%): (□) 0.64% Pt/γ-Al_2O_3; (○) 5% $CuCr_2O_4$/γ-Al_2O_3.

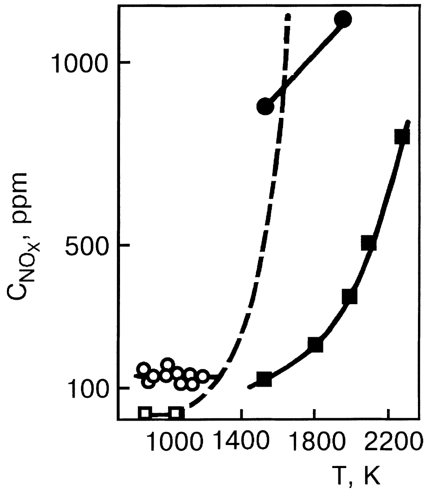

Fig. 4. Dependence of NO_x concentration on temperature during flame combustion ((●) fuel NO_x; (■) thermal NO_x)) and catalytic combustion in CHG ((○) fuel NO_x; (□) thermal NO_x)), N_f=1 wt%. The broken line corresponds to equilibrium values of NO_x concentration.

gen to NO_x can be decreased below 5–10% as compared to 40–100% with flame combustion. Thus, the catalytic combustion in CHG has been shown to provide effective control of both thermal and fuel NO_x (Fig. 4).

4. Main applications of fluidized bed catalytic combustion

This method of fluidized bed catalytic combustion was tested in pilot and industrial plants in various technological processes: (1) heating and evaporation of liquids; (2) drying and thermal treatment of powder materials; (3) destruction of organic wastes; (4) treatment of waste-waters containing organic admixtures; (5) treatment of waste-water sediments and sludges.

4.1. *Heating installations*

Various water-heating devices of different capacity working on gas and liquid fuels were designed and tested. They showed high efficiency of fuel utilization and provided environmentally clean fuel combustion. The commercial production of water-heating boilers of a capacity of 230 kW was started in Novosibirsk under the license of the Boreskov Institute of Catalysis. They are used in industry for local heating of production sites. The recommended fuels are diesel fuel and fuel oil, but other liquid fuels, e.g. residual oil or spent hydrocarbon solvents, can also be used. CHGs are manufactured as both stationary and mobile heat sources. The heating efficiency of these boilers is ca. 90% and they produce water of a temperature of 95°C as a heating agent. Mobile heat supply units were proved to be very efficient and convenient for heating buildings under construction in winter time as temporary heat sources.

4.2. *Installations for drying and thermal treatment of powder materials*

In these installations powder materials to be treated are fed directly into the upper part of a fluidized catalyst bed and the heat produced in fuel combustion is used for their treatment. The size of powder particles is less than that of catalyst granules, and the treated material is carried out of the catalyst bed with the exhaust gases and captured in a cyclone. The direct contact (solid–solid) of the powder material with the catalyst granules results in a very high rate of the drying process. Processes for drying coal and phosphogypsum were tested in prototype installations and showed very high efficiency. Drying of coal slurry was carried out in an installation of 3.5 MW heat capacity producing 20 t/h of dried coal. This method has shown the following advantages in comparison with conventional methods of drying based on the use of heat produced in flame combustion: decrease of emission of pollutants, safety against fire and explosion, decreased size and weight of the installation, and saving of fuel.

Another example is pulsed thermal treatment of gibbsite in CHG producing amorphous alumina – a product with unique properties (primarily a high reactivity) permitting to convert it to aluminum hydroxide with pseudoboehmite structure, which is a starting material for preparation of alumina adsorbents and catalyst supports [13]. This technology is commercialized since 1988 in Kazakhstan.

4.3. Incineration of liquid organic wastes

On prototype installations numerous studies have been performed with the aim to determine the applicability of the process of catalytic fluidized bed combustion to the destruction of a wide range of industrial organic wastes. The experiments were made with used organic solvents: gasoline, acetone, cyclohexane, ethanol, xylene, toluene, butylacetate, styrene contaminated with paints and lacquers; wastes of polyester and epoxy resins; amines and isocyanates; wastes of petroleum products free of mineral admixtures. The incineration of wastes proceeded under stable regimes, and without soot formation. The combustion efficiency exceeded 99.9%. The concentration of carbon monoxide in flue gases was below 0.01 vol%, and that of thermal nitrogen oxides below 5 ppm [20]. The catalyst normally deactivated during first 5–10 h of operation, but later its characteristics (activity, mechanical strength, specific area) were practically unchanged during hundreds of hours of operation. The catalytic activity was retained at a level sufficient for complete waste oxidation. Several installations for organic liquid waste destruction with utilization of heat were built and put into operation at industrial enterprises in Russia and Ukraine.

4.4. Treatment of waste-waters containing organic admixtures

The method of fluidized bed catalytic combustion allows to treat waste-waters containing organic admixtures. The process is autothermal when the concentration of organics is 10–15%, at larger organics concentration excess heat is utilized, e.g. by production of process steam. This process was tested in pilot and industrial installations with a capacity of up to 2 m^3 of waste-water per hour. The following advantages were established:

- suppression of formation of nitrogen oxides and products of incomplete combustion;
- no expensive refractory materials are required for construction of installations due to low temperatures;
- fire- and explosion-safety;
- saving of fuel.

4.5. Treatment of waste-water sediments and sludges

The method of catalytic fluidized bed combustion was shown to be very effective in the treatment of waste-water containing organic sediments, e.g. in paper and pulp industry. When these sludges are burned in a catalytic heat generator the organic part is oxidized completely, and the mineral part is captured in a cyclone. The resulting materials have a developed porous structure and a large surface area, and can be used as efficient and inexpensive adsorbents for purification of waste waters. This technology was implemented for the combined treatment of waste-waters and waste-water sediments (pulps) at the paper and pulp complex "Grigishkes", Lithuania [40].

The project on the study of catalytic destruction of agricultural sludges and manure was recently started. This problem is of great importance in Europe, because such wastes spread bad odors and cause pollution of air, soil and water with toxic substances. A special pilot installation consisting of a fluidized bed catalytic reactor and downstream gas cleaning units was designed. The installation was assembled at the testing site and now it is being prepared for start up and testing. The tests of the installation will include a treatability study with different types of organic sludges and wastes, optimization of operational regimes and study of the effect of the reaction media on the operational characteristics of the catalyst: BET area, pore structure, activity, chemical and phase composition, and mechanical strength.

5. Application of fluidized bed catalytic combustion in demilitarization technologies

5.1. Catalytic destruction of mixed organic waste containing radionuclides

At nuclear material processing plants there are large quantities of accumulated mixed organic wastes of complex composition containing vacuum pump and industrial oils, used extractants and their solvents and various hazardous organic compounds contaminated with uranium, plutonium and their fission products. These wastes represent a threat to the environment upon their storage and treatment because of the combination of inflammability, explosiveness, toxicity and radioactivity. Due to the absence of safe treatment technologies, these wastes are kept in tanks. Sometimes they were treated by unacceptable methods, e.g., by open incineration, but now this is prohibited. The use of flame combustion leads to the formation of toxic nitrogen oxides and other pollutants and generation of sub-micron radioactive particles, which may be emitted into the atmosphere with combustion off-gases, and refractory type plutonium dioxide, which is hard to process in the aqueous recovery process.

On the basis of the experience accumulated at BIC (Boreskov Institute of Catalysis) we proposed to treat such wastes in a fluidized catalyst bed reactor. The application of this technology would result in a decrease of pollutant emission and formation of low-temperature fired ash, which is easier to capture and process.

The Boreskov Institute of Catalysis together with Lawrence Livermore National Laboratory (USA) and a number of nuclear material processing plants and institutions in Russia and abroad proposed to the International Science and Technology Center (ISTC) in Moscow a project entitled “Development of Technology of Catalytic Fluidized Bed Destruction of Mixed Organic Wastes Containing Radionuclides”. The project was approved by the Russian Ministry of Atomic Energy, evaluated as “excellent” by a number of Western experts and accepted for funding since 1 November 1995 by the ISTC Governing Board [41–44].

The main results accomplished during the first year of activities are the following.

The inventory of mixed organic wastes of Siberian nuclear materials processing plants according to waste amounts and compositions was carried out.

Two experimental installations of 1 kg/h capacity were constructed in Novosibirsk. One, at the Plant of Chemical Concentrates, was built for the experiments with radioactive wastes of the Plant to study catalyst performance in destruction of real radioactive materials. The experiments were carried out with wastes of lubricants and extractants and showed high efficiency of waste destruction and stable operation.

Another installation at NGPII VNIPIET was constructed for the experiments with simulated non-radioactive wastes. It was specially designed for the study of exhaust gas purification from dust of surrogate non-radioactive materials and acid gases. It includes catalytic reactor, cyclone, scrubber, two absorber-condensers and aerosol filter.

The optimization of the processes of catalytic combustion and exhaust gas purification on these installations was carried out. Under optimum conditions the concentrations of pollutants in exhaust gases were: NO_x – 5–15 ppm, CO – 1–20 ppm, SO_2 – 1–10 ppm, non-radioactive dust – below 0.1 mg/m^3.

The construction of the demonstration plant at the Plant of Chemical Concentrates and its testing was planned in 1997 (Fig. 5).

The successful accomplishment of this project, on the one hand, would form the basis for future industrial applications of this technology at other nuclear material processing plants in Russia and abroad, and on the other hand, would help to solve the urgent environmental problem of the Novosibirsk region.

This technology will find other applications, for example in the elimination of hazardous wastes from the chemical industry (such as pesticides), and possibly, of special significance, destruction of the components of chemical weapons.

5.2. Prospects of fluidized bed catalytic combustion for destruction of chemical warfare agents

The committee on Alternative Chemical Demilitarization Technologies of the US National Research Council collected and studied information on alternative and supplemental technologies for the destruction of components of chemical weapons. Among the processes considered for possible research and devel-

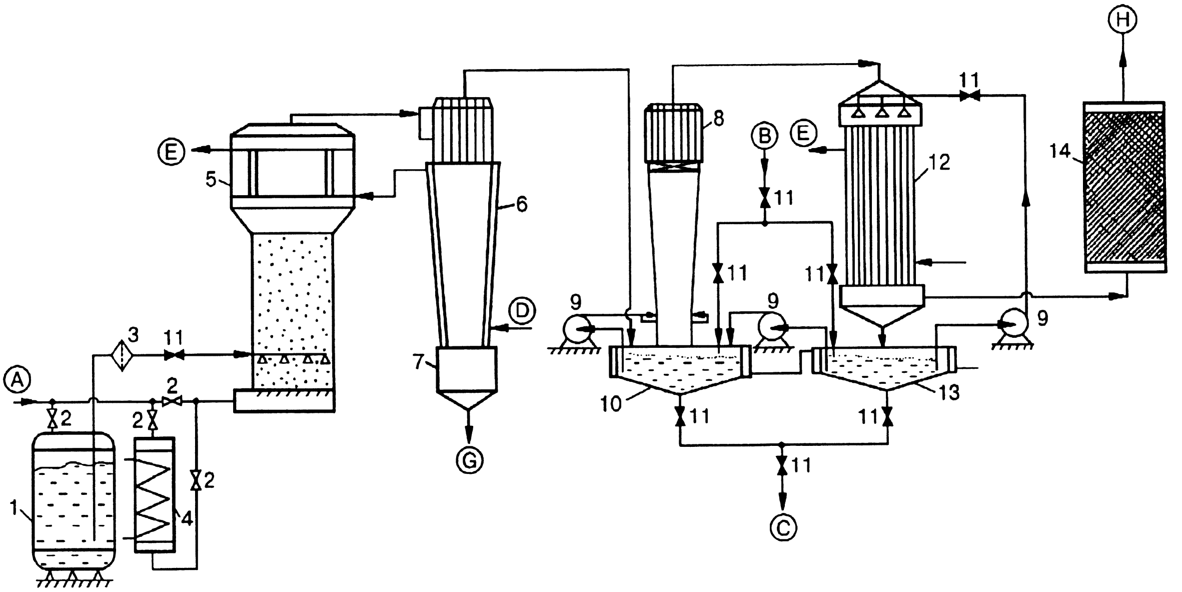

Fig. 5. Principal scheme of the demonstration plant for the treatment of mixed organic radioactive waste in a fluidized catalyst bed: 1 – tank with waste, 2 – gas flow regulators, 3 – filter, 4 – electric heater, 5 – catalytic reactor, 6 – cyclone, 7 – collector, 8 – scrubber, 9 – pump, 10, 13 – tanks, 11 – valves, 12 – absorber–condenser and 14 – aerosol filter; A – supply of compressed air, B – supply of water, C – removal of spent scrubber liquids, D – cooling water inlet, E – cooling water outlet, G – ash removal and H – purified exhaust.

opment is fluidized bed oxidation [45]. The following key development factors are mentioned:

- choose catalyst for controlling acid gases, improve destruction and removal efficiencies;
- identify solid handling equipment for removing spent catalyst and providing fresh makeup;
- characterize all effluents streams.

The promising technological schemes are staged processes when chemical agents are initially oxidized in a catalytic fluidized bed reactor and the exhaust gases are purified from toxic compounds either by scrubbing with neutralizing solutions or by the treatment in a molten salt bed [46].

Because of extremely rigid requirements imposed on technologies for destruction of chemical warfare agents (at least 99.9999% destruction efficiency) the implementation of the fluidized catalytic bed technique in this field is a very difficult problem, but its application in the near future for destruction of less hazardous substances, e.g. pesticides prohibited for use, toxic rocket fuels and propellants, e.g. asymmetric *N*,*N*-dimethylhydrazine, seems to be conceivable.

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