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(71) **Applicant:** UNIWERSYTET JAGIELLONSKI [PL/PL];
ul. Golebia 24, PL-3 1-007 Krakow (PL).

(72) **Inventors: NAJBAR, Mieczysława**; ul. Lużycka 57/45, PL-30-060 Krakow (PL). **LECH, Ryszard**; ul. Grzepskiego 20, PL-30-698 Krakow (PL). **DANIELEWSKI, Marek**; Łazy 106, PL-32-048 Jerzmanowice (PL). **BUDZIOCH, Janusz**; ul. Rozrywka 20/9, PL-3 1-4 17 Krakow (PL).

(74) **Agent:** LUBNICKA, Aleksandra; Uniwersytet Jagiellonski - Ciotru, ul. Czapskich 4, PL-3 1-1 10 Krakow (PL).

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(54) Title: THE METHOD OF SIMULTANEOUS REMOVAL OF NO AND CARBON PARTICLES AND INORGANIC DUST FROM FLUE GASES AND CATALYTIC REACTOR FOR REMOVAL OF NO AND CARBON PARTICLES AND INORGANIC DUST FROM FLUE GASES

(57) Abstract: The method of simultaneous removal of NO and carbonic particles and inorganic dust from flue gases in the reactor equipped with the catalyst for direct decomposition of nitric oxide located on a metallic monolith consists in tangential introduction of flue gases to the reactor circumference generating rotational flow of the flue gases downwards of the reactor with simultaneous flow disturbance due to flue gases contact with undulating surface of metallic foil located on an inner wall of the reactor chamber and split of the flue gases by contact with the catalyst located on a spiral band falling to the lower part of the reactor, and next flue gases jet direction counter-currently to a cylindrical inner chamber containing the slices of the monolithic catalyst disturbing laminar flow of the flue gases jet. The deposited solid particles of the pollutants are collected in the lower part of the reactor. The invention concerns also the reactor designed for simultaneous removal of NO and carbon particles and inorganic dust from flue gases.

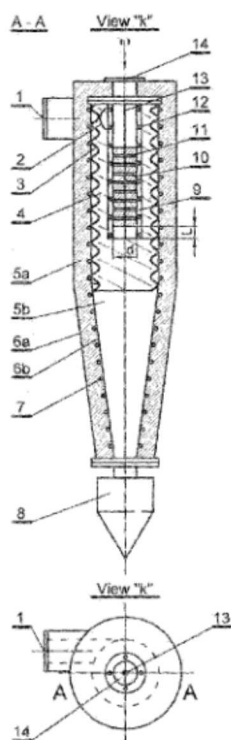


Fig. 1

TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW,
ML, MR, NE, SN, TD, TG).

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The method of simultaneous removal of NO and carbon particles and inorganic dust from flue gases and catalytic reactor for removal of NO and carbon particles and inorganic dust from flue gases

The subject of the invention is the method of nitric oxide (NO) removal from flue gases containing oxygen and the reactor for removal of NO, first of all from dusty flue gases which are generated by the stationary emission sources.

Hitherto the removal of NO from the dusty flue gases is carried out in this way, that the flue gases containing NO flow axially to the inlets of the channels of a monolithic catalyst and the flow is laminar. The catalyst is prepared using a ceramics, fire-resistant material or metal foil and it is covered by catalytic active substance. During laminar flow through the channels of the monolithic catalyst the flue gases come to contact with the catalyst and the chemical reactions occur. Reduction of NO to N_2 is the result of these reactions. Simultaneously the solid particles deposit on the surface of the monolithic catalyst and it is necessary to remove those particles periodically using a mechanical methods.

A catalytic reactor for cleaning the dust-free flue gases in which cleaning of gases occur almost evenly in central and outer part of the metallic monolithic catalyst is well-known from the description of the Japanese patent No. 2006-196988. In the appliance, a magnetic substance and catalytically active substance are contained in a layers created on surface of the channels of a metallic monolith. The monolith is cylindrical or elliptical or quadrangular and it is made out of undulating austenitic stainless steel. On the outer side of the monolithic catalyst a stable magnet is placed or a magnetic field generator. The monolithic catalyst is located in a casing, which is made of non-magnetic material with high thermal resistance. The casing is located between the stable magnet or the magnetic field generator and the monolithic catalyst, which is enveloped by a ceramic fibre mat. The mat is a thermal insulator of heat transported from cleaned gases to the stable magnet or the magnetic field generator. The cleaned flue gases are directed to an inlet cross-section of the monolithic catalyst using a flue gas channel. The geometric axis of the flue gas channel and the geometric axis of the monolithic catalyst overlap. A diffuser is installed at the end of the flue gas channel and it is a coupler between the flue gases channel and the metallic casing of the monolithic catalyst.

The flue gases are cleaned as a result of a chemical reaction proceeding on the catalyst during their flow through the channels of the monolithic catalyst created from an undulating metallic foil.

The aim of the invention is the method and the reactor for the direct removal of nitric oxide from the dusty flue gases allowing for simultaneous efficient decomposition of nitric oxide and systematic removal of solid particles, especially carbonic particles.

The method of the flue gases cleaning according to the invention consists in simultaneous removal of NO and the solid particles from the flue gases as well as in the possible combustion of the carbon particles carried by the flue gases. Those processes proceed due to repeated contacts of the flue gases and the dust with the surfaces containing the active catalyst. The contacts are ensured by the changes of the flue gases jet flow direction in the reactor.

The removal of nitric oxide and the solid particles from the dusty flue gases according to the invention is performed in the reactor with the catalyst for direct decomposition of nitric oxide deposited on metallic monolith. According to the method of the invention the dusty flue gases enter the reactor tangentially to a circumference. It causes a rotational flow of the flue gases downwards. The flow undergoes disturbance because of the contact of the flue gases jet with undulating surface of the metallic foil placed on an internal wall of the reactor chamber and a split of the flue gases jet due to contact with a spiral band of the foil. The surface of the metallic foil placed on internal wall of reactor chamber as well as the surface of the spiral band are covered by the active component of the catalyst. Afterwards flue gases jet is directed countercurrently to a cylindrical inner chamber, in which the slices of the monolithic catalyst are installed and the laminar flow of the flue gases is disturbed. In the course of the cleaning process the falling solid particles are accumulated on a bottom of the reactor.

The preferred disturbance of flue gases laminar flow in the inner chamber of the reactor is achieved by spacers present between parallel slices of the monolithic catalyst in the reactor or by nonparallel location of the slices of the monolithic catalyst.

Preferably, the porous slices of the monolithic catalyst with variable porosity are used. The preferred temperature in the reactor is in the range of 150°C - 450°C. Preferably an oxide catalyst made by direct oxidation of the foil, especially of the acid-proof austenitic steel foil is used. The catalyst in the most favourable form contains the phase with $\alpha\text{Fe}_2\text{O}_3$ structure

and the phases with spinel structure having the lattice parameters closed to those of NiFe_2O_4 or the spinel phases only. The phases create microcrystallinities, which contain Cr and Mn additionally and possibly also Si (Polish patent application P.395905). Using this kind of a catalyst it's possible to remove simultaneously the nitric oxide, the solid particles and to combust the carbon particles contained in the dust transported in the flue gases jet.

The catalytic reactor for removal of nitric oxide and the carbon particles and the inorganic dust from dusty flue gases according to the invention comprises the catalyst on the metallic support for the direct removal of nitric oxide. The reactor is at least partially cylindrical and is equipped with a thermal insulation and its inlet is located in an upper part in such a way, that the flue gases are introduced tangentially to the reactor circumference. The upper part contains a chamber with an undulating inner surface, which is covered by an active catalyst phase. The spiral band made of acid-proof steel foil and covered by the active phase of the catalyst is located inside of the chamber with the undulating inner surface and moreover the spiral band falls downwards. There is an inner chamber heated with an inner heater in the geometric axis of the chamber with the undulating inner surface. The slices of the monolithic catalyst are installed in that chamber. The reactor is equipped with a casing containing two cylindrical coaxial walls with an insulating material between them. A casing heater adjoins at least to a part of the inner wall of the casing. In the lower part of the reactor a tight closure is located and it is simultaneously a dust container.

Preferably, the catalyst is also located on the outer surface of the inner chamber.

It is possible to locate slices of the catalyst parallel towards each other and vertically towards the reactor axis. In this case the directing spacers are installed between the slices of the monolith catalyst. The shape of the directing spacers induces the rotational flow of the flue gas jet. The most preferred is the shape of a propeller or similar to the shape of the propeller.

If the front of the monolithic slices of the catalyst is located in such a way, that the ratio of the distance L measured from the inlet to the heated chamber of the slices of the monolithic catalyst to the inner diameter d of the heated chamber of the slices of the monolithic catalyst $L/d > 50$, then the directing spacer is being set in front of the inlet to the slice of the monolithic catalyst. But if the ratio $L/d < 50$, then the directing spacer is not being set before inlet to the porous support of the catalyst.

It is possible to situate the slices of the monolithic catalyst at an angle towards other slices and towards the axis of the reactor.

Preferably, there is an additional heater in axis of the heated chamber of the slices of the monolithic catalyst located.

Preferably, the reactor has a cylindrical shape in the part, where the cylindrical chamber with undulating surface is located, and a conical shape in the remaining part. The chamber with undulating surface may be in contact with the internal wall of the reactor casing.

The method and the reactor according to the invention are designed especially for the removal of NO and carbonic particles and inorganic dust from flue gases created in the stationary emission sources.

Multi-stage process of the decomposition of nitric oxide and the removal of the solid particles occurs in the reactor according to the invention. Flue gases in the reactor come to contact with the catalyst located on the metallic surfaces. They simultaneously direct the flow of flue gases, making a countercurrent arrangement of the jets in the reactor. The first stage of the catalytic reactor is the chamber with undulating inner surface, the second one is the spiral band. The spiral band forms a spiral flow of flue gases around its geometrical axis, directing flue gases downwards of the reactor, besides the catalytic performance. The change of the direction of flue gases flow causes removal of the solid particles from the cleaned flue gases jet and increases an inlet effect in the slices of the monolithic catalyst. Removal of the dust containing the solid particles from flue gases jet is caused by the flow of the gaseous jet according to a curved trajectory. Dust from the flue gases comes to the contact with the catalyst located on the surface of the first stage of the multistage catalytic reactor and while it is falling down under influence of the gravitational force, it is being removed from the flue gases jet and moreover the carbonic particles contained in the dust are totally oxidized due to the activity of the catalyst, if the catalyst contained in the reactor enables the oxidation process.

The flue gases jet with lower contents of the solid particles flows to the heated chamber of the slices of the monolithic catalyst. The inlet to the heated chamber of the slices of the monolithic catalyst, which is next stage of the multistage catalytic reactor is located in the lower part of the cylindrical chamber with undulating inner surface and the spiral band in such a way, that the cleaned flue gases change the flow direction turning back to the heated chamber of the slices of the monolithic catalyst and countercurrently come to the reactor inlet being already cleaned in a significant degree from the dust contained in the inlet flue gases of the reactor. In the inner chamber of the multistage catalytic reactor the slices of the

monolithic catalyst are installed and the cleaned flue gases flow through their channels. Gas flowing in countercurrent continuously comes into the contact with the catalyst causing its cleaning, but to make the contact easier and to increase a purity degree of the flue gases, the flow of the flue gases jet in the last stage of the multistage catalytic reactor is subjected to often changes of the direction, forcing the cleaned flue gases jet into an alternating rotational or rectilinear flow. The change of the direction of the flow is achieved by means of the directing spacers or due to location of the slices of the monolithic catalyst at an angle towards the plane of the cross-section of the reactor. The directing spacer is never located on the outlet of the last slice of the monolithic reactor. Flow of the cleaned gas through the slices of the monolithic catalyst depends on the shape and area of the cross-section of their channels, which are variable in an preferred version. The change of the direction of the flow of cleaned flue intensifies contact between dusty gas and the catalyst. The dust removed from flue gases jet is collected in a tight closure of the reactor and it is periodically removed from the tight closure of the reactor. During normal exploitation of the reactor the tight closure does not allow for an entrainment of the air from environment.

The method and the reactor according to the invention enable intensive, continuous contact of flue gases with highly developed surface of the catalyst and assure changeability of the flue gases flow direction. The contact of the flue gases with the catalyst located on the surface of the channels is facilitated due to very often changes of the flow direction of flue gases simultaneously enabling of the removal of dust from the flue gases jet.

The examples of reactor structure according to the invention are shown on the schematic drawings. A longitudinal-section and cross-section of the reactor are shown in fig. 1 for the ratio $L/d < 50$. The longitudinal-section of the reactor for ratio $L/d > 50$ is shown in fig. 2. The spiral band, which is performed in one part is shown on fig. 3. The spiral band, which is prepared in separate parts is shown in fig. 4. The directing spacer is shown in fig. 5. The slices of the monolithic catalyst installed under the various angles are shown in fig. 6. The slices of the monolithic catalyst situated parallel towards each other are shown in fig. 7. And the slices of the monolithic catalyst with various sizes of their cross-section are shown in fig. 8.

The example of the reactor structure according to the invention is made out for ratio of the dimensions $L/d < 50$. The dusty gas generating in a stable emission source is supplied using feeding channel 1 which is thermally insulated from environment impact and the gas is introduced using the dusty gas inlet 2 tangentially to the surface of the cylindrical chamber

with the undulating inner surface 3 for removal of NO and solid particles as well as for the oxidation of carbonic particles presented in dust transported by the gaseous jet. The catalyst enabling the removal of NO from the gaseous mixture and oxidation of carbonic particles occurs on the undulating cylindrical surface prepared using acid-proof austenitic steel foil. The oxide phases created on the acid-proof austenitic steel foil by oxidation are liable to arise as the catalyst. Inside the cylindrical and undulating chamber 3 the spiral band 4 made of the austenitic acid-proof steel covered by the oxide phases is located and it is falling downwards of the chamber 3 and it is the support of the catalyst for removal of NO and for the oxidation of carbonic particles contained in the dust. The spiral band may be prepared as one part or it may be prepared from separate parts creating the band, as it is shown on fig. 4, but distances between the individual band parts should not overlap according to the band pitch. The spiral band 4 is stretched out on the band frame 15. The cylindrical and undulating reactor chamber 3 is placed in the casing composed of the cylindrical part 5a and the conical part 5b. The casing contains thermal insulation, e.g. a light heat-resistant insulating material, and the insulation is placed between the two coaxial walls: the outer casing wall 6a and the inner casing wall 6b but the heater 7 of the casing is in touch with the inner casing wall 6b. The conical part 5b of the reactor in its lower part is equipped with the tight closure 8, which is simultaneously a periodically cleaned dust container. In the geometrical axis of the cylindrical undulating chamber 3 a heated chamber of the slices of the monolith catalyst 9 is placed and its surface are covered by the active component of the catalyst. In the heated chamber of the slices of the monolith catalyst 9 the slices of the monolithic catalyst 10 and/or 16 are located and they are of different area of the channel cross-sections. The slices of the monolithic catalyst 10 and/or 16 are covered by the same active component of the catalyst as the cylindrical chamber of the reactor with undulating surface 3 and the spiral band 4.

If the ratio $L/d < 50$ the directing spacer 11, shown on fig. 5, is not situated in the front of the inlet to porous support of the catalyst 10 and/or 16. The directing spacer 11 consists of the blades 17, which are sloped at small angle towards level and fastened between the outer ring 18 and the inner ring 19. On the wall of heated chamber of the slices of the monolithic catalyst 9 the chamber heater 12 is installed. In the geometrical axis of the heated chamber of the slices of the monolithic catalyst 9 an additional axial heater 13 is located. The heated chamber of the slices of the monolithic catalyst 9 is equipped with the gaseous outlet 14. The slices of the monolithic catalyst 10 may be placed under the various angles α (alpha) or β (beta) towards the axis of the heated chamber of the slices of the monolith catalyst 9 as

it is shown in fig. 6, in order to attain a change of the flow direction for achieving an effect of inlet flow to the slice of the monolith catalyst. At the time the directing spacers 11 are not applied. The channels 20 in slice of the monolithic catalyst are shown in fig. 6. The change of the flow direction before the inlet to the slices of the monolithic catalyst is achieved when the slices of the monolithic reactor are parallel towards themselves, but under angle α (alpha) towards the axis of the heated chamber of the monolithic slices of the catalyst 9, as it is shown in fig. 7. There is a space filled by the flue gases jets between the slices of the monolithic catalyst 10 with both arrangement of the slices of the monolith catalyst 10 shown in figs. 6 and 7.

The example of the structure of the reactor according to the invention shown in fig. 2 is designed for the case of the dimensions ratio $L/d > 50$. The difference in the example of the structure shown in fig. 2 in comparison with the structure shown in fig. 1 is in the location of the directing spacer 11 in the front of the inlet to the porous slice of the monolithic catalyst 10 or ii, when the front of the porous support of the catalyst 10 is in position that the ratio $L/d > 50$.

In both the cases of the realization the length of the heated chamber of the slices of the monolithic catalyst 9 should be as long as possible in order to achieve maximum distance for the flow of the flue gases.

It is possible to apply the slices of the monolithic catalyst 16 with the area cross-sections of the channels different from the area cross-sections of the channels of the slices of the monolithic catalyst 10, that is seen in fig. 8.

The slices of the monolithic catalyst 10 are of the same or the various shapes and dimensions of the channels cross-sections and each chamber may contain the slices of the monolithic catalyst with the various shapes and cross-sections.

NOTATION

- 1 - feeding channel
- 2 - dusty gas inlet
- 3 - cylindrical chamber of reactor with undulating inner surface
- 4 - spiral band
- 5a - cylindrical part of casing
- 5b - conical part of casing
- 6a - outer casing wall
- 6b - inner casing wall
- 7 - heater
- 8 - tight closure
- 9 - heated chamber of the monolith slices of catalyst
- 10 - slice of monolithic catalyst
- 11 - directing spacer
- 12 - chamber heater
- 13 - axial heater
- 14 - gas outlet
- 15 - band frame
- 16 - slice of monolith catalyst with variable shape and dimensions of cross - section of the channels
- 17 - blade
- 18 - outer ring
- 19 - inner ring
- 20 - channel of slice of a monolithic catalyst

CLAIMS:

1. The method of simultaneous removal of NO and carbon particles and inorganic dust from flue gases in the reactor equipped with a catalyst for direct decomposition of nitric oxide located on a metallic monolith characterised in that the flue gases flow tangentially to the reactor circumference generating rotational flow of flue gases downwards of the reactor with simultaneous disturbance of the flow due to contact of the flue gases jet with undulating surface of metallic foil covered by the active component of the catalyst, located circumferentially on the inner wall of the reactor chamber, and split of the flue gases jet due to contact with the catalyst deposited on a spiral band falling to the lower part of the reactor, and next the flue gases jet is directed countercurrently to a cylindrical inner chamber consisting of the slices of the monolithic catalyst and laminar flow of the flue gases jet is disturbed, but the deposited solid particles of the pollutants are removed from lower part of the reactor.
2. The method according to the claim 1, characterized in that the disturbance of the flue gases laminar flow in the reactor inner chamber is achieved by placing the directing spacers between parallel slices of the monolithic catalyst.
3. The method according to the claim 1, characterized in that the disturbance of flue gases laminar flow in the reactor inner chamber is achieved by nonparallel arrangement of the slices, of the monolith reactor in relation to each other.
4. The method according to the claim 1, characterized in that the slices of the monolithic catalyst with various shapes and dimensions of the channels cross-section are applied.
5. The method according to the claim 1 characterized in that the temperature inside the reactor is maintained in the range of 150°C - 450°C.
6. The method according to the claim 1, characterized in that the oxide catalyst on metallic base is applied.
7. The method according to the claim 6, characterized in that the oxide catalyst on base of acid-proof austenitic steel is applied.
8. The method according to the claim 1, characterized in that the slices of the monolithic catalyst contain the oxide phases with $\alpha\text{Fe}_2\text{O}_3$ structure and with spinel structure of the lattice parameters closed to NiFe_2O_4 or the spinel phases only, but these phases create microcrystallites, which contain Cr and Mn additionally and possibly Si.

9. The method according to the claim 1 or 8, characterized in that the simultaneous removal of nitric oxide and total oxidation of carbonic particles contained in the dust transported by the flue gases proceeds.
10. The method according to the claim 1, characterized in that the removal of nitric oxide originating from the stationary emission sources occurs.
11. The catalytic reactor for removal of NO and carbonic particles and inorganic dust from flue gases, containing the slices of the monolithic catalyst for direct removal of nitric oxide having at least partially cylindrical shape with thermal insulation, with inlet located in the upper part, characterized in that the inlet generates tangential introduction of dusty gas into the reactor and the reactor contains the cylindrical chamber with undulating inner surface 3, with an active phase of the catalyst, located in the upper part of the reactor and inside the chamber 3 with undulating inner surface a spiral band 4 falling towards lower part with active component of the catalyst on its surface is located, whereas in the geometric axis of the chamber 3 with undulating surface a chamber 9 heated by a heater 12 is situated and the slices of the monolithic catalyst 10 and/or 16 are located in the heated chamber 9, moreover the reactor is equipped with a casing with two coaxial walls 6a and 6b and a heat resistant material is placed between the walls 6a and 6b, but a heater 7 is in touch, at least partially, with the wall of the inner casing 6b while in the lower part of the reactor a tight closure 8 is located and it is simultaneously a dust container.
12. The reactor according to the claim 11, characterized in that the active component of the catalyst is present on the inner wall of the chamber 9.
13. The reactor according to the claim 11, characterized in that the slices of the monolithic catalyst are parallel towards themselves and vertical to axis of the reactor.
14. The reactor according to the claim 11 or 13, characterized in that the directing spacers 11 are located between the slices of the monolithic reactor.
15. The reactor according to the claim 14, characterized in that the shape of the directing spacers 11 forces rotational flow of flue gases jet.
16. The reactor according to the claim 15, characterized in that the shape of the directing spacers 11 is similar to a propeller.
17. The reactor according to the claim 11, characterized in that the heater 13 is located in the axis of the chamber 9.

18. The reactor according to the claim 11, characterized in that the directing spacer is located in front of the inlet to the monolith slices of the catalyst when the front of the monolithic slices of the catalyst is in the position that the ratio $L/d > 50$, but when the ratio $L/d < 50$ the directing spacer 11 is not situated before the inlet to the slices of the monolithic catalyst.
19. The reactor according to the claim 11, characterized in that the slices of the monolithic catalyst are positioned at angle to each other and towards the axis of the reactor.
20. The reactor according to the claim 11, characterized in that the inner wall of the casing 6b is in touch with the chamber 3.
21. The reactor according to the claim 11, characterized in that it is composed of the cylindrical part in which the chamber 3 is located and the conical remaining part.
22. The reactor according to the claim 11, characterized in that it contains the oxide catalyst on metallic base.
23. The reactor according to the claim 11 or 22, characterized in that the oxide catalyst is deposited on acid-proof austenitic steel base.
24. The reactor according to the claim 23, characterized in that the catalyst contains a phase with $\alpha\text{Fe}_2\text{O}_3$ structure and a phase with spinel structure and the lattice parameters closed to NiFe_2O_4 or the spinel phases only, but the phases create microcrystallinities, which contain Cr and Mn additionally and possibly Si.
25. The reactor according to the claim 11, characterized in that it is to be used for the simultaneous removal of nitric oxide from the flue gases, oxidation of carbonic particles contained in dust transported by flue gases and removal of inorganic dust.
26. The reactor according to the claim 11, characterized in that it is to be used for removal of nitric oxide generated by the stationary emission sources.

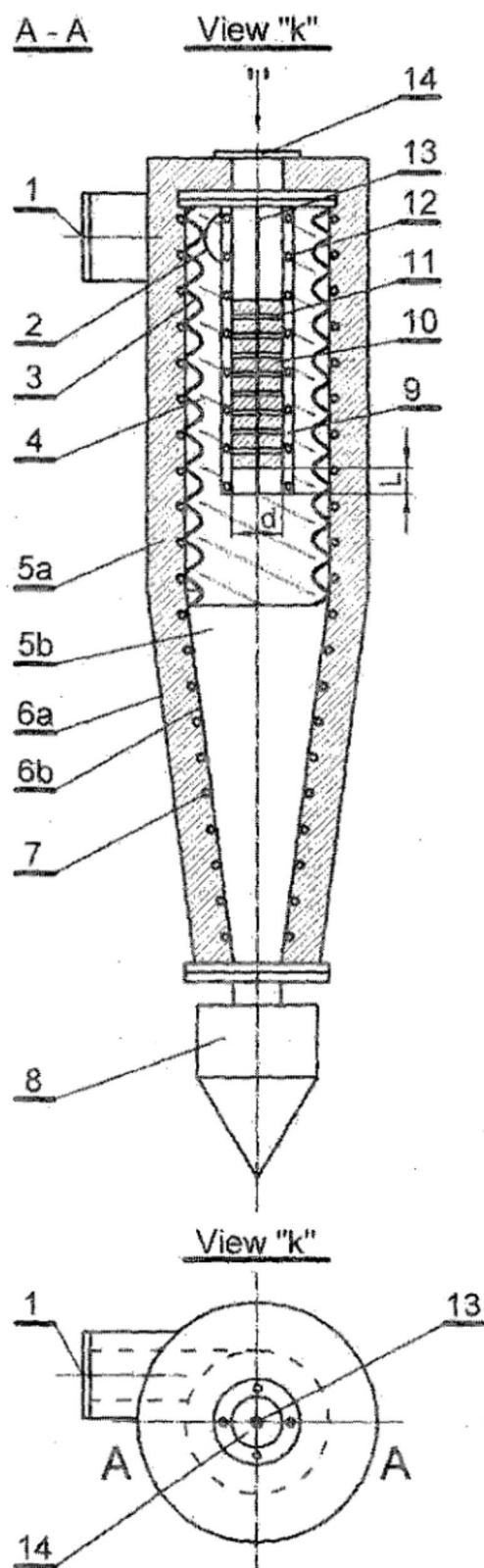


Fig. 1

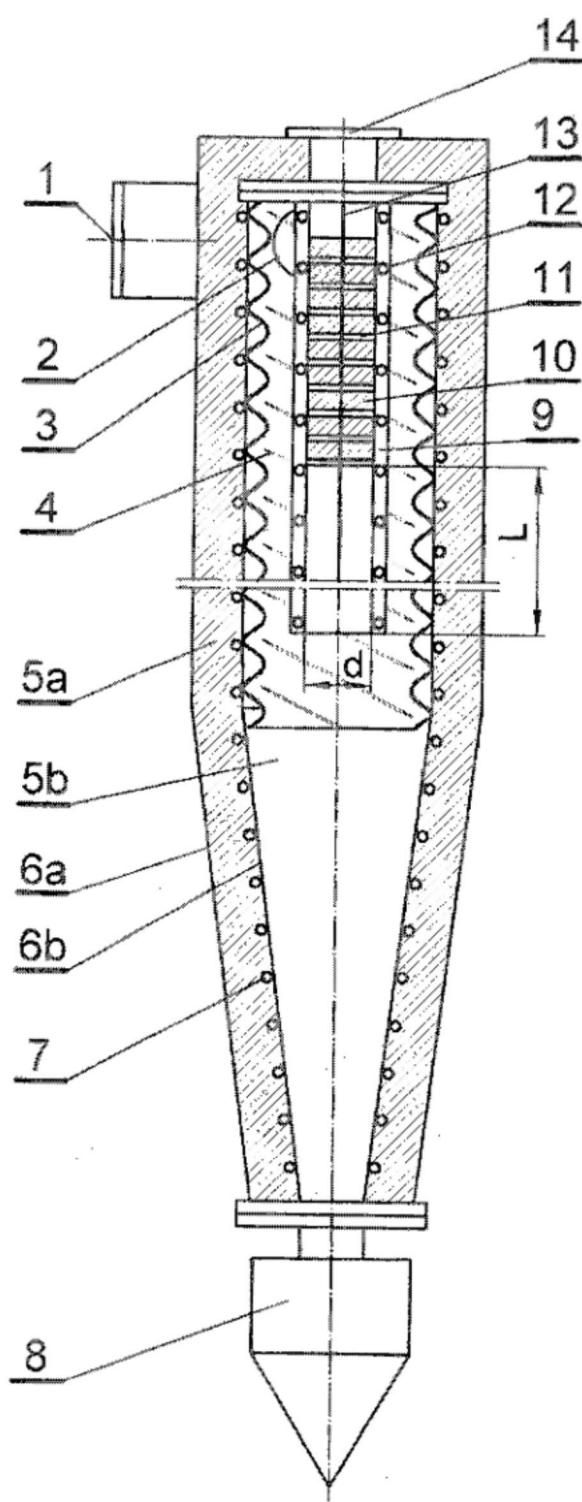


Fig. 2

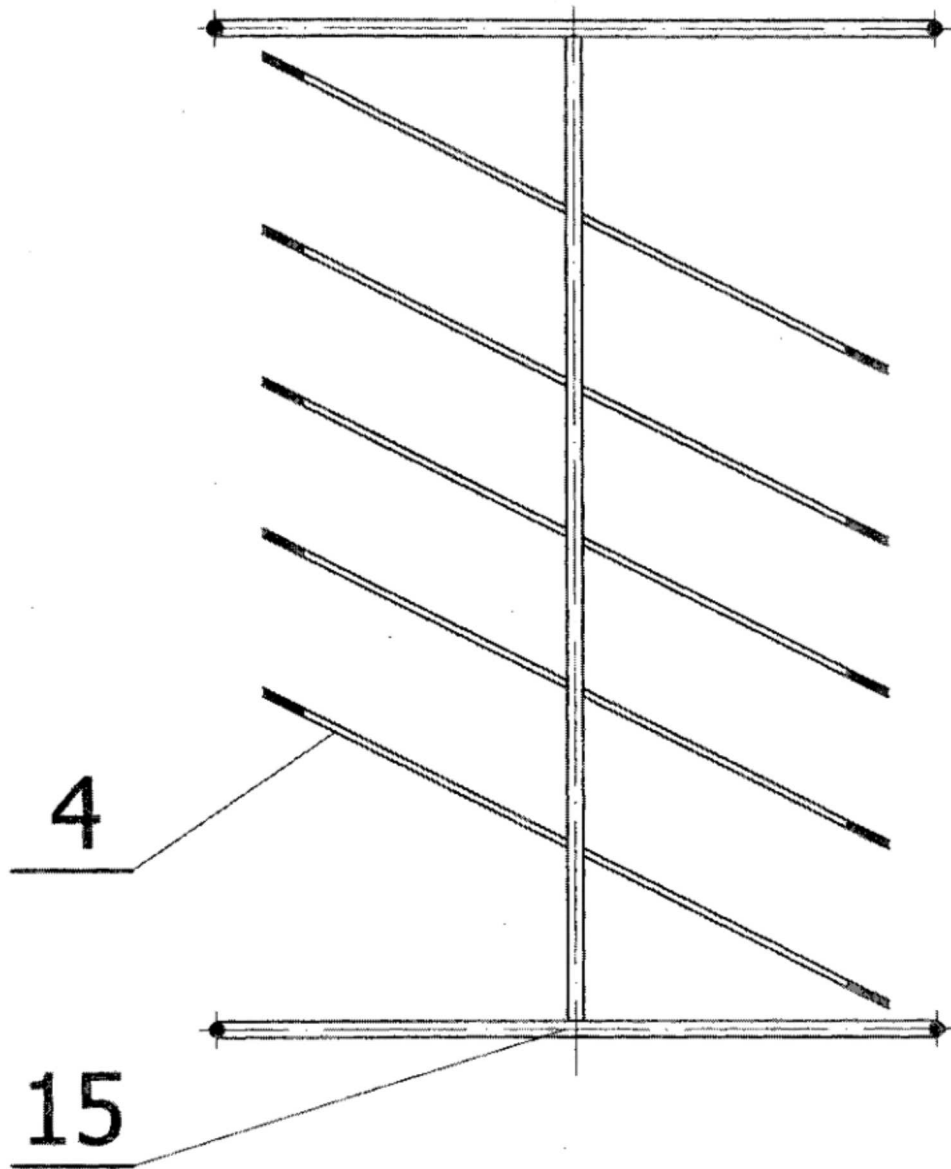


Fig. 3

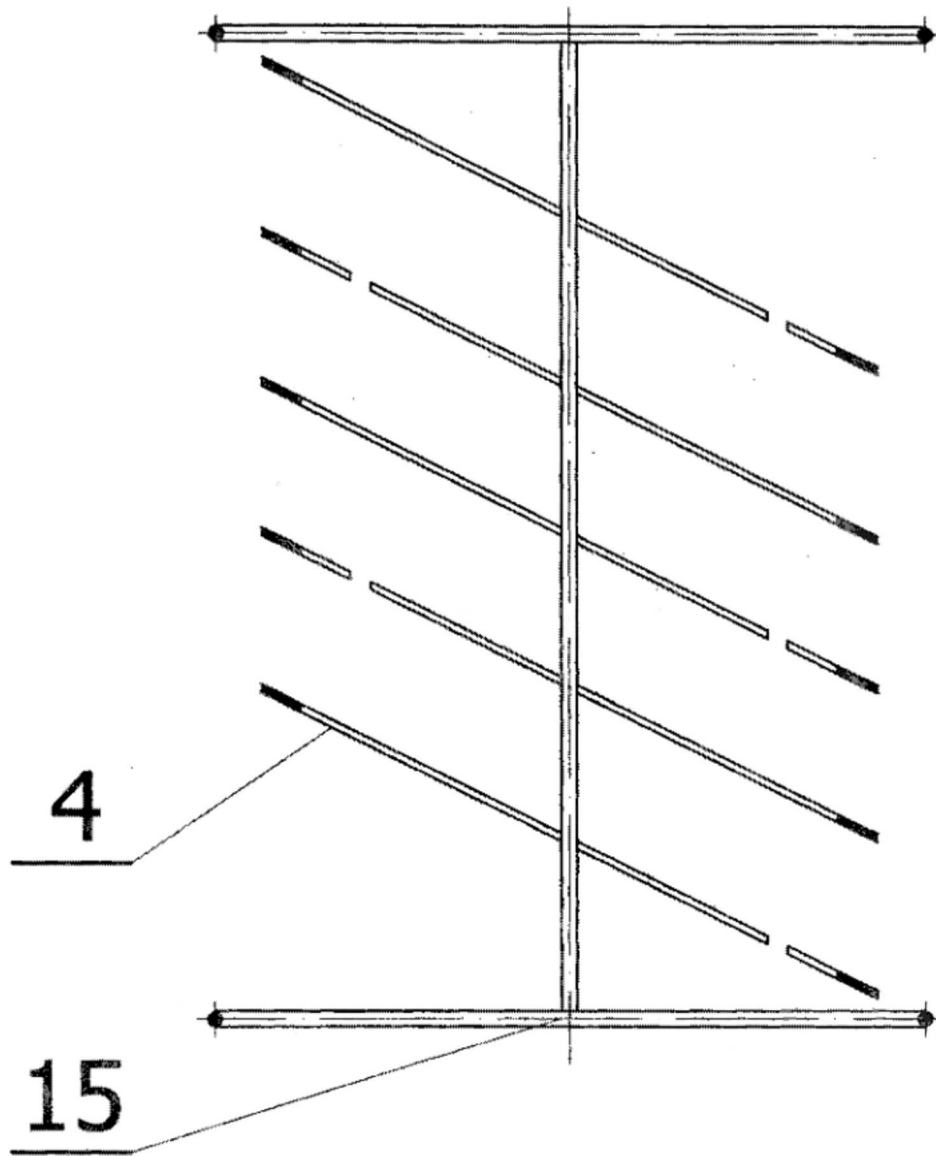


Fig. 4

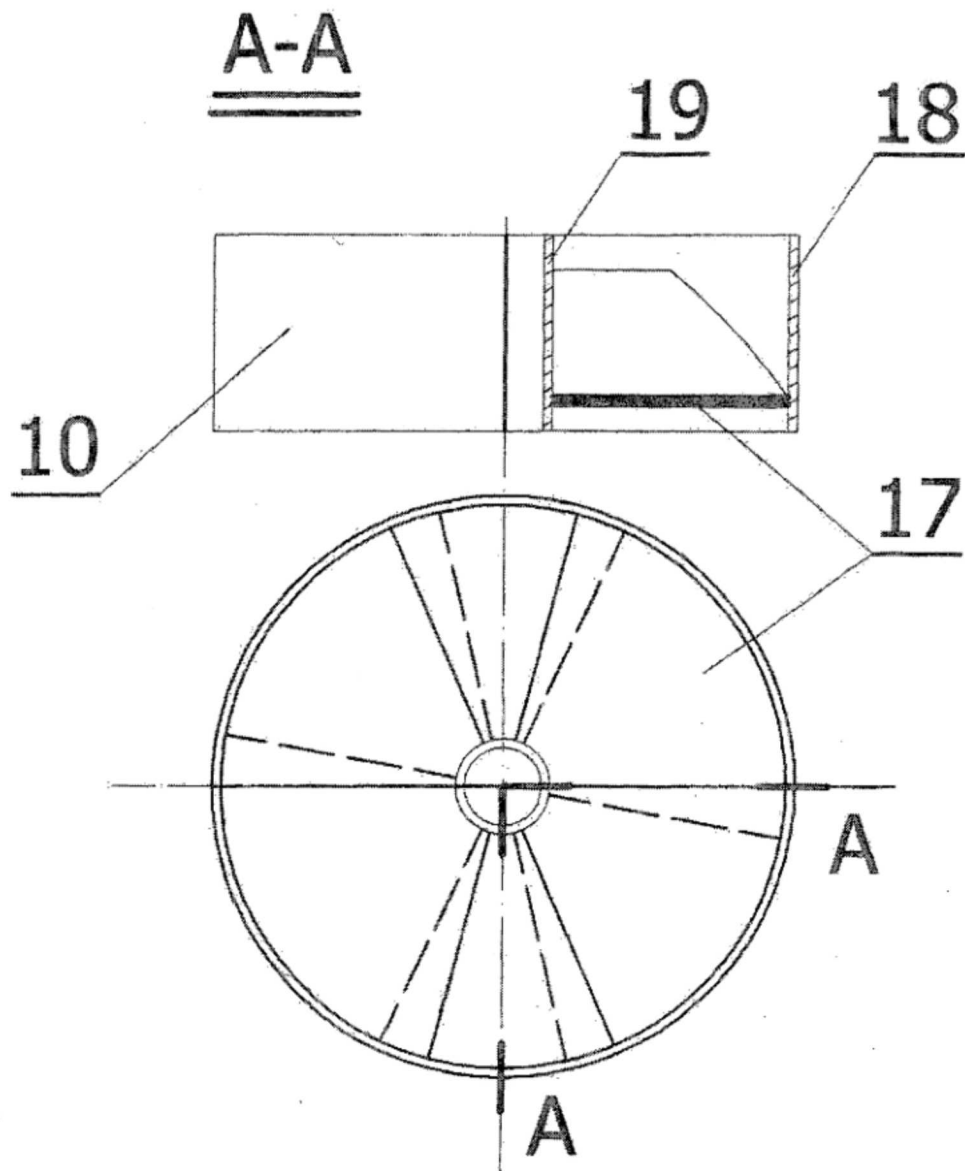


Fig.5

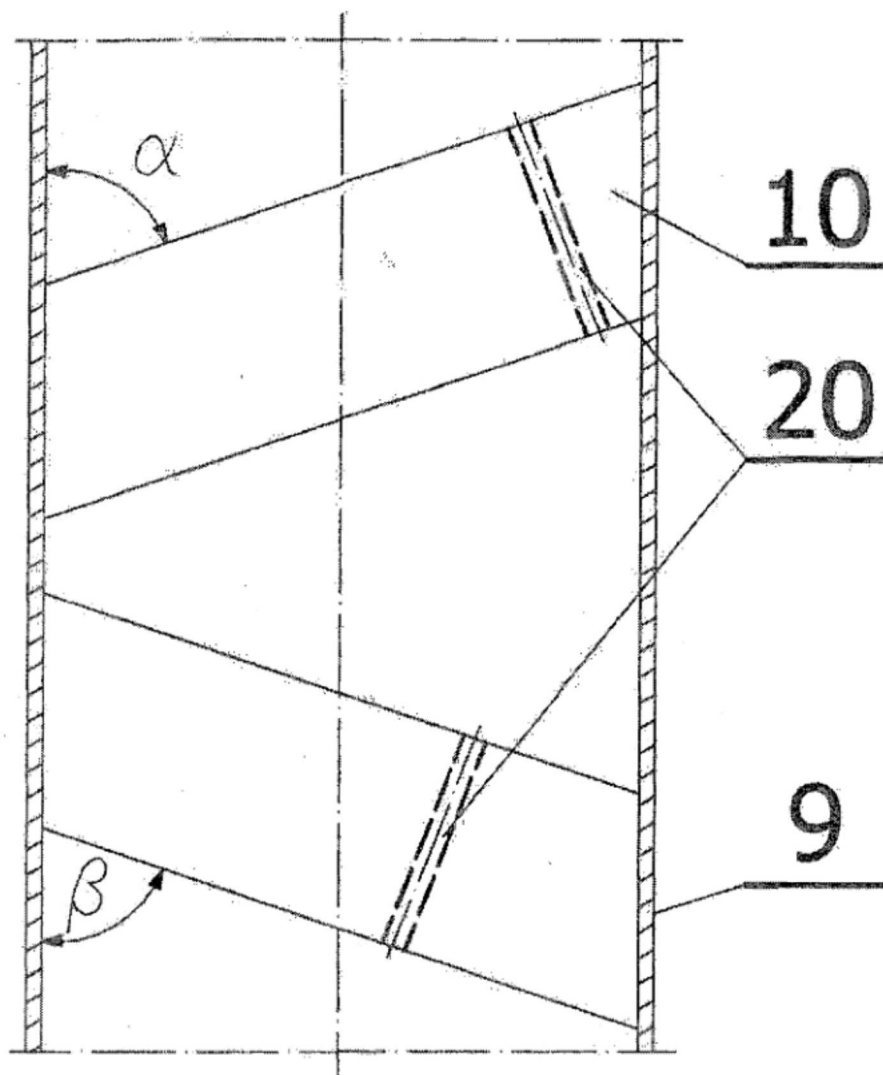


Fig. 6

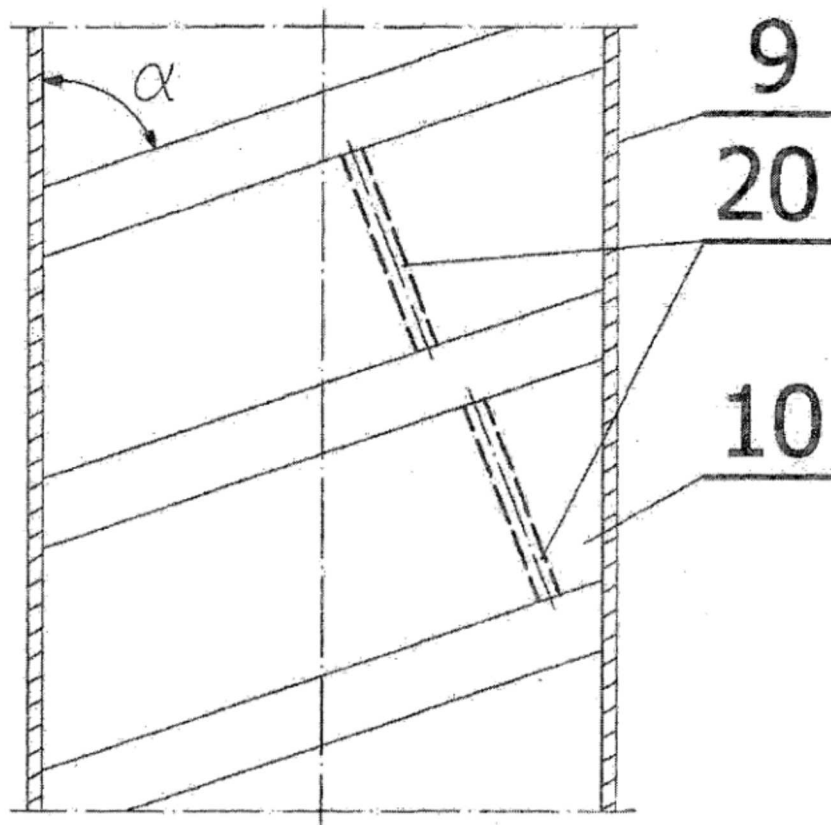


Fig. 7

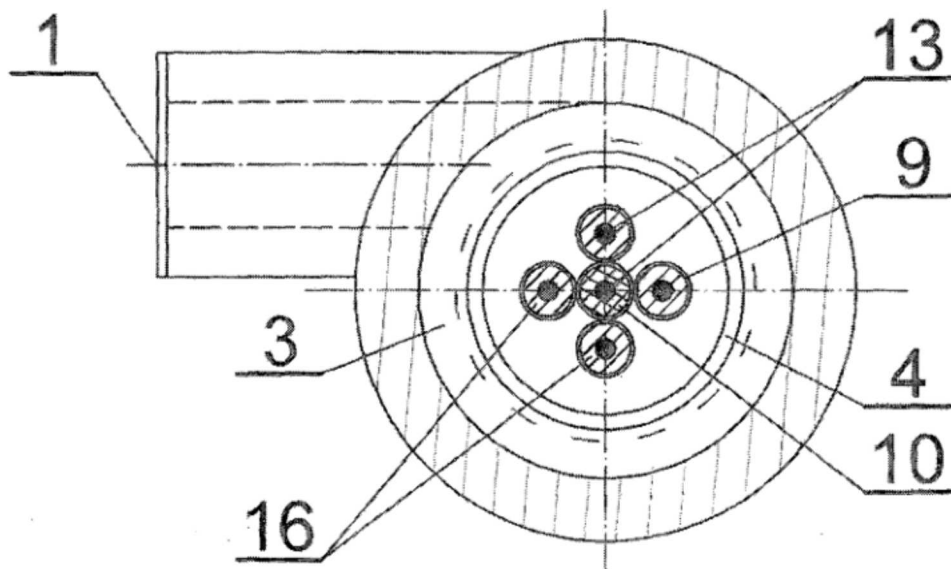


Fig. 8

INTERNATIONAL SEARCH REPORT

International application No

PCT/PL2012/000129

A. CLASSIFICATION OF SUBJECT MATTER

INV. B01D53/86

ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal , WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 2 311 549 A1 (MITSUBISHI HEAVY IND LTD [JP]) 20 April 2011 (2011-04-20) -----	1, 11
A	US 4 834 962 A (LUDWIG GERHARD [DE]) 30 May 1989 (1989-05-30) -----	1, 11



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INTERNATIONAL SEARCH REPORT

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International application No

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