

Method for Evaluating and/or Controlling a Process in a Vessel for Unit Operations

The present invention relates to a method for evaluating and/or controlling a process in a vessel for unit operations involving an electrically conductive bed, e.g., a bed containing electrically conductive particles.

5 Unit operations may involve a chemical conversion and/or a physical transformation. A given process may require one or more unit operations to obtain the desired product from a starting material or feedstock.

For example, hydrogen can be produced from hydrocarbon fuels by non-oxidative conversion which includes thermal decomposition of hydrocarbons into hydrogen and carbon, also referred to as dissociation or pyrolysis. Carbon is produced as a valuable byproduct of the process. The reaction can be run by employing electrically heated moving beds of carbon particulates maintained at the reaction temperature. The carbon
10 particulates are heated by an electrical potential or voltage applied across at least a portion of the particle bed. The resistance of the discrete yet interconnecting electrically conductive particulates to electrical current results in the particulates being heated.

Moving bed reactors, while representing a great improvement over the previous methods used in the art, still suffer from serious operational problems. When the fluid hydrocarbon stream contacts the hot
15 particulates, it is cracked, and carbonaceous material, such as coke, is formed. Most of this newly formed material will deposit on the surface of the moving particulates in the particle bed. However, the cohesive nature of the material may give rise to formation of agglomerates of carbon particles that may result in plugs. Typically, the agglomerates/plugs travel at a slower flow rate than the loose solid particles in the reactor bed. The plugs can continue to grow and may ultimately lead to clogging or other reactor failures.

20 Uniform temperature distribution over the cross-section as well as uniform velocity distribution of the particulates are important prerequisites to ensure the intended function and optimum reaction control of the moving bed reactor. Hence, it is desirable to detect anomalies in the temperature and velocity distribution in a quick and efficient manner. It is important for critical operating conditions in the reactor, which can lead to clogging, to be detected and eliminated at an early stage. This improves the availability of the reactor.
25 Accumulation of undesirable coke deposits should be detected early and eliminated by appropriate measures. This maximizes reactor run time between scheduled shutdowns. In addition, operating conditions of the reactor can be safely set near critical operating conditions. This maximizes reactor performance.

In the state of the art, there are no known methods to measure temperature distribution as well as particle velocity distributions in an appropriate temporal and spatial resolution.

30 The installation of probes or sensors within the moving bed is impractical for several reasons. Stationary installed sensors in the moving bed impair the flow of solids and can provoke uneven distributions of, e.g., velocity and temperature. The moving bed exerts considerable forces on stationary internals. Consequently, there is a risk that stationary installed sensors are bent or torn off, which may cause further damage of the reactor and downstream installations. Analogous problems are encountered in a heated fixed bed during
35 the filling and draining of the reactor. In a fluidized bed probes or sensors can be damaged by abrasion from the bed material. Additionally, considerable forces act on installations that protrude far from the reactor wall into the interior of the reactor. The detection of electrical quantities inside a moving bed, a fixed bed or a fluidized bed through which electric current is flowing is impaired due to the lack of galvanic isolation of electrical sensors from the bed.

Hence, there is a need for methods for contactless evaluating and/or controlling of processes in a vessel for unit operations involving a bed containing electrically conductive particles.

Some approaches for implementing contactless inductive measurement techniques are already known.

5 Griffiths in Meas. Sci. Technol. 12 (2001) 1126–1131, Magnetic induction tomography describes magnetic induction tomography (MIT) as a new imaging modality being developed for the process industry and for medical imaging. MIT applies a magnetic field from an excitation coil to induce eddy currents in the material to be studied, and the magnetic field from these is then detected by sensing coils.

10 Hampel et al. in Sensors 2022, 22, 2309, A review on fast tomographic imaging techniques and their potential application in industrial process control provide an overview of the current state of the art in fast process tomography and its potential for use in industry.

Sieger et al. in tm–Technisches Messen 2023; aop: <https://doi.org/10.1515/teme-2022-0103>, Kontaktlose induktive Strömungstomografie in grundlegender und angewandter Fluidodynamik describe Contactless Inductive Flow Tomography (CIFT) as a flow measurement technique that can reconstruct the global 3D flow in electrically conducting fluids.

15 Hauer et al in Journal of Power Sources 2005, 143, pp. 67-74, Magnetotomography—a new method for analysing fuel cell performance and quality describes a method to visualize the 2D distribution of current between the two electrodes of a fuel cell. The method is also patented (DE 10 2006 035 741 A1).

20 Morgan in ACS Catal. 2016, 6, 1356–1381, Evolution and Enabling Capabilities of Spatially Resolved Techniques discuss and evaluate the development of spatially resolved techniques using electromagnetic and physical probes to investigate the progression of the chemistry within a catalyst bed.

WO 2023/225438 A1 describes a system comprising a reaction chamber, the reaction chamber having a cylindrical shape and containing a catalyst; a fluidization plate having two sides, a first side of the fluidization plate being tangibly connected to a first end of the reaction chamber; a gas input receiver tangibly connected to a second side of the fluidization plate; at least one output mechanism tangibly connected to a second end of the reaction chamber; and an inductive coil encircling the reaction chamber, wherein during the operation of the system the inductive coil creates an electromagnetic field. The inductive coil may be controlled by an oscillator, and the system may also use sensors in communication with a control system. The sensors are said to be able to provide information to the control system regarding, e.g., the temperature, pressure, flow rates of gases, magnetic flux, electrical field intensity, efficiency of the process, how much catalyst remains in the catalyst unit, how much output has been generated, and how much output gas is leaving the system.

35 The present invention is intended to solve the problem described as follows: An electric current is injected in an electrically conductive bed, e.g., a bed of electrically conductive particles. In certain embodiments, the electric current concurrently provides electric resistance heating to the reactor bed. Typically, the volume between the electrically conductive particles is filled with gas, the electrical conductivity of which can be neglected in comparison to the particles. For this reason, the electric current is channeled only through the particles. This electric current generates a magnetic field that can be measured outside the bed. A change in size or any rearrangement of the particles inevitably changes the electric current distribution and thus also the associated magnetic field. The magnetic field data therefore conveys information about the material distribution in the reactor. Deviations from a uniform particle distribution or defects such as inclusions, voids,

cracks or the like, can be localized and their propagation velocity in the reactor can be determined. The temperature distribution is also closely linked to the electrical current distribution, i.e. high current densities lead to a local increase in temperature.

5 The invention relates to a method for evaluating and/or controlling a process in a vessel for unit operations involving an electrically conductive bed, comprising

- a) applying a voltage across at least a portion of the bed through a plurality of electrodes that are in an electrically conductive relationship with the bed to induce an electric current flowing through the bed; and
- 10 b) measuring a magnetic field and/or measuring changes in a magnetic field induced by the electric current by magnetic sensors at a plurality of locations outside the bed to generate magnetic field data.

In an embodiment, the method additionally comprises

- c) establishing electric field characteristics, including the topology and/or the dynamics of the electric field in the bed, using the magnetic field data, and
- 15 d) calculating a distribution of matter in the bed and/or distribution of electrical conductivity in the bed using the electric field characteristics established in c).

Establishing electric field characteristics from the magnetic field data will also be referred to as "reconstructing the electric field" herein.

In a further embodiment, the method additionally comprises

- 20 e) adjusting one or more manipulated variables of the unit operation according to the distribution of matter in the bed and/or distribution of electrical conductivity in the bed calculated in d).

The bed can contain a regular or a random packing. Particular embodiments of the packing are described for example in [Eigenberger, G., & Ruppel, W. (2000). Catalytic fixed-bed reactors. Ullmann's Encyclopedia of Industrial Chemistry]. A regular packing may advantageously comprise honeycomb monoliths, foam blocks or corrugated plates, preferably honeycomb monoliths. A random packing (hereinafter: "particle bed")
25 contains a large number of loose particles. The particles can have a regular shape, for example spherical, cylindric, ring-shaped, or an irregular shape, for example split.

The term "evaluating" is used herein to refer to any type of detecting, assessing or analyzing, and may be qualitative or quantitative. Evaluating may include imaging, i.e., visualization of the process. The term "controlling" means that manipulated variables of the unit operation are adjusted according to the evaluation.

30 The process of the invention is distinguished from previously known Magnetic Induction Tomography (MIT) and Contactless Inductive Flow Tomography (CIFT). Unlike MIT where an electric current is induced in the volume to be monitored by generating an excitation field using an induction coil, an electric current is injected directly into an electrically conductive bed by applying a voltage thereto. In the case of CIFT, an electric current density distribution in a fluid to be measured is generated by the interaction of the velocity distribution
35 and an exciting magnetic field, which is produced by an induction coil.

The method of the invention may be used, e.g., to evaluate a process with regard to the occurrence of inhomogeneities in the particle bed, such as formation of agglomerates or void spaces, as well as surveillance of the progress and homogeneity of drying or calcination operations, and the like.

5 Generally, a fluid stream, which is typically a gaseous stream, is flowing through the bed. The fluid stream may serve different purposes. In one embodiment, the fluid stream carries reactants, e.g. gaseous reactants, over the bed and/or carries reaction products, e.g., gaseous reaction products, out of the bed. In another embodiment, the fluid stream carries vapors, such as moisture or solvent vapors, out of the bed.

10 The vessel is preferably an elongated vessel, preferably a vertical elongated vessel. Vessels having an essentially circular cross section are generally preferred. In embodiments, the voltage is applied along the vertical axis of the vessel such that the electric current induced in the bed flows substantially parallel to the vertical axis of the vessel. As will be appreciated, at positions where there are inhomogeneities in the bed, the electric current can also have a field component normal to the general direction of current flow.

Alternatively, the voltage may be applied radially across the center of the bed to a perimeter of the bed.

15 The unit operation may be selected from chemical conversions such as dehydrations and dehydrogenations; and physical transformations such as drying operations. Vessels in which chemical conversions are performed are typically referred to as reactors. The terms "vessel" and "reactor" are thus used interchangeably herein without, however, necessarily implying a limitation to chemical conversions.

The particle bed may be a moving particle bed, a fluidized particle bed or a fixed particle bed.

20 The method employing a moving particle bed typically comprises feeding the particles to the reactor at the top of the particle bed, allowing the particles to slide downwardly in the gravitational field as a continuous column, and withdrawing the particles from the reactor at the bottom of the particle bed. Discharging the particles may be accomplished by conventional discharge means, e.g., by a rotary valve.

The downward particle velocity suitably may be in the range of 0.1 to 50 m/h, preferably 0.2 to 20 m/h, more preferably 0.5 to 10 m/h.

25 The method employing a fluidized particle bed typically comprises fluidizing the particles by an upwardly flowing fluidizing gas injected at the bottom of the reactor to form a fluidized bed of particles.

In the case of one or more gaseous reactants, these may be introduced together with the fluidizing gas and may even constitute the fluidizing gas. Gaseous reaction products will simply be carried out by the fluidizing gas.

30 In a fixed-bed reactor, the electrically conductive particles are stationary and do not move with respect to the reactor.

According to the invention, the bed is electrically conductive, such that an electric current can flow through the bed. When the bed is a regular structure, it can be intrinsically electrically conductive, or be coated with an electrically conductive coating.

35 In a particle bed, the particles comprise electrically conductive particles through which an electric current flows. The particle bed must contain a sufficient proportion of electrically conductive particles to ensure

macroscopic electrical conductivity of the particle bed and allow for the passage of an electrical current through the bed. The proportion of electrically conductive particles in the particle bed can be in the range between 5% and 100% , preferably between 20% and 100 % , most preferably between 50 and 100% by volume. In any event, the conductivity, and the nature of the conductive particles constituting the particle bed, will depend largely on the process being carried out.

Electrically conductive particles may consist of a material which is inherently conductive or of a material which may be rendered conductive by the reactive deposition of an electrically conducting material, e.g., carbon, or by the adsorption or absorption of a polar liquid such as water.

In one embodiment, the particle bed contains chemically active particles. Chemically active particles include catalytically active particles, reactants of a chemical conversion or carriers for deposition of products of the chemical conversion. Catalytically active particles exert a catalytic activity on the chemical conversion. The chemically active particles may be reactants or products of the chemical conversion, particularly in a moving bed or fluidized bed method.

The electrically conductive particles themselves may be chemically active or may be essentially catalytically inert.

The electrically conductive particles may be admixed with non-conductive particles. The non-conductive particles may be chemically active particles.

Suitably, the electrically conductive particles are selected from carbonaceous particles, silicon carbide particles, metallic particles, or composites thereof. The electrically conductive particles may be in the form of fines, or be agglomerated such as pelletized or granulated, if required.

In another embodiment, the electrically conductive particles are selected from adsorbents and absorbents, such as superabsorbent polymers, comprising varying degrees of moisture.

Carbonaceous particles may include particles of char or coke, in particular graphite particles. Metallic particles, may be metallic throughout or have a metallic coating over a non-metallic core. Metallic particles include conductive prereduced oxidic particles.

The unit operations of the present method can either be exothermic or endothermic. In the case of endothermic unit operations, the supply of heat is required. The method of supplying heat is not particularly restricted and different concepts can be used. The heat source can be combustion heating or electric heating.

Preferably, the heating means envisaged in the present invention are electrical heating means. The use of electric energy as a heat source instead of heating by combustion of natural gas allows considerable advantages, in particular with regard to the ease of control. The use of electricity offers opportunities for the use of compact, modular, high performance and energy efficient reactors. When the electricity derives from a non-fossil resource, the endothermic unit operation can be implemented without net emission of carbon dioxide. The heating energy can be generated directly in the bed or in separate heating elements.

In one embodiment, the electric current concurrently provides electric resistance heating to the unit operation. This means the electric current passed through the electrically conductive bed generates thermal energy.

In another embodiment, heat can be provided by inductive heating.

- 5 If the electrical current is applied as AC, a frequency is typically applied that is distinguishable from the power line frequency, with a difference of, for example, 10 Hz.

- In yet another embodiment, indirect resistive heating elements may be used. The indirect resistive heating elements can take the shape of a wire, ribbon, sheet or strip and can be straight, meandering or coiled. Such a heating element converts electricity into heat through the process of Joule heating. The electric current used for indirect heating also generates a magnetic field. However, this magnetic field does not contain any usable information about the process, as this current does not flow through the bed and is therefore not influenced by the conditions there. The contribution of the magnetic field from indirect heating is compensated during data processing, or the electric current flowing through the indirect resistive heating elements is switched off when and as long as the magnetic field induced by the electric current flowing through the bed is measured, in order to avoid interference or distortion of the result.
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In another embodiment, heat can be provided to the unit operation by preheating a fluid that is designed to flow through the bed, e.g., a gaseous reactant stream.

The different concepts of supplying heat may be combined. For example, the thermal energy generated by passing the current through the electrically conductive bed may be supplemented by other heat sources.

- 20 In the method of the invention, a voltage is applied across at least a portion of the bed to induce an electric current flowing through the bed. The electrical power is supplied through a plurality of electrodes that are in an electrically conductive relationship with the bed, e.g., immersed in the particle bed or electrically connected to the regular structure.

- The electrodes may be made of a metal, graphite or any other suitably conductive material. Various composite refractory materials, for example containing graphite or metal, may be employed. Electrical power may be supplied through a plurality of electrodes that are in an electrically conductive relationship with the bed, e.g., immersed in the particle bed.
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- Preferably, a pair of axially spaced electrodes is employed, i.e., upper and lower electrodes. Alternatively, a radial electrode arrangement may be contemplated with a central electrode extending along the axis of the reactor and a generally cylindric counter electrode or a pair of concentric cylindrical electrodes with a cylindrical inner electrode and a cylindrical outer electrode so that an electric current is induced to flow through the bed in a radial direction between electrodes.
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- The shape of the electrodes is not particularly limited. The measuring principle can cope with virtually any type of electrodes such that the design of the electrodes has no significant impact on its performance. However, information about the electrode design is generally taken into account as a boundary condition in reconstructing the electric field from the magnetic field data. Information about the electrode design may also be taken into account in determining the locations of magnetic sensors outside the bed to identify an optimal arrangement of the magnetic sensors.
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Preferably, the electrodes are designed to achieve a uniform current distribution within the particle bed. Preferably, the electrodes take the shape of a grid or of rods. Suitable electrode assemblies are described in WO 2019/145279 A1.

5 When rods are used, each electrode preferably comprises a number of rods distributed across the cross section of the particle bed. Electrode rods that run to a point are particularly advantageous. Preferably, the upper and lower electrode rods run to a point on the side toward the heated zone. The tip may be conical or wedge-shaped. Correspondingly, the end of the rod may take the form of a dot or a line. The rod electrodes are connected to the hood in an electrically conductive manner and are jointly supplied with electrical power via the hood.

10 Preferably, the electrodes take the form of a grid. For grid form, various configuration variants are conceivable, for example grids in honeycomb form composed of advantageously regular polygons, rectangular grids formed from parallel bars, grids in the form of spokes or grids composed of concentric rings. Particular preference is given to grids in the form of spokes and grids composed of concentric rings.

15 The potential or voltage may arise from an A.C. or D.C. source. Advantageously, a potential difference (voltage) of 1 volt to 10 000 volts, preferably of 10 volts to 5000 volts, more preferably of 50 volts to 1000 volts, is applied. The electrical field strength between the hoods is advantageously in the range of 1 V/m to 100 000 V/m, preferably 10 V/m to 10 000 V/m, more preferably 20 V/m to 5000 V/m, especially 30 V/m to 1000 V/m.

20 The specific electrical conductivity of the bed is advantageously from 0.001 S/cm to 60 MS/m, preferably from 1 MS/m to 50 MS/m (e.g. metallic catalysator) and from 0.01 S/cm to 300 S/cm, especially from 0.1 S/cm to 100 S/cm (carbon bed).

25 This advantageously results in an electric current density in the bed of 0.01 A/cm² to 100 A/cm² and higher, preferably from 0.05 A/cm² to 50 A/cm², especially from 0.1 A/cm² to 10 A/cm². Generally, the lower limit of the electric current density is given by the distance of the magnetic field sensor to the bed resulting in magnetic field changes in the order of 1 nT or higher due to the inhomogeneities in the bed. There is no upper limit, so that 1000 A/cm² and 10000 A/cm² and higher might also be possible.

The electric field traveling through the bed induces a magnetic field. The magnetic field is illustrated in Fig. 1. If the electric current flows predominantly in the vertical direction, a magnetic field is formed with field lines mainly in the azimuthal direction.

30 According to the invention, the magnetic field induced by the electric current is measured by magnetic sensors at a plurality of locations outside the bed. In order to safeguard the magnetic sensors from contamination and for easy access to the sensors, the magnetic sensors are generally positioned at the outside of the reactor shell. In certain cases however, magnetic sensors can also be located inside the reactor shell, depending on the shape of the reactor. For example, magnetic sensors can be located inside
35 the reactor shell between a refractory lining and the reactor shell.

Hence, when magnetic sensors are said to be positioned at the circumference of the bed, this includes arrangements wherein the magnetic sensors are located inside the reactor shell between a refractory lining and the reactor shell, in particular between a refractory lining and the reactor shell, as well as arrangements wherein the magnetic sensors are located outside of the reactor shell. Likewise

- Where the magnetic sensors are positioned at the outside of the reactor shell, the material comprising the reactor shell should not substantially affect the magnetic fields emanating from the bed through which the electric current is traveling. Hence, the reactor shell is constructed from non-magnetic materials. It is preferred that a physically strong, low electric conductivity, non-corrosive and non-magnetic material, like titanium or austenitic stainless steel, is used. A shell of non-magnetic austenitic stainless steel is generally preferred. Generally, a lining of refractory material electrically and thermally insulates the bed from the reactor shell. Suitable refractory materials include aluminum oxide, zirconium oxide and mixed oxides of aluminum, magnesium, chromium, silicon, as well as concrete. The refractory lining is typically provided in the form of bricks. The magnetic property of the material is described by the relative magnetic permeability μ_r . Non-magnetic materials are characterized by $\mu_r = 1$ as ceramics, titanium and copper, while austenitic steels like the heavily used grades 1.4301 or 1.4401 have $\mu_r < 1.3$. Steel grades with $\mu_r < 1.1$ are preferable. The class of non-magnetic steels is given by the Stahl-Eisen-Werkstoffblättern (SEW) 390. A specific heat treatment can reduce the magnetic permeability after milling. For the process of the invention, the magnetic permeability of the reactor shell should be in the range of $1 \leq \mu_r \leq 1.3$.
- Suitably, the magnetic sensors may be positioned at the circumference of the reactor and/or different planes along the axis of the reactor.

The electrodes and magnetic sensors may have one of the following configurations:

- a) the plurality of electrodes comprise a pair of axially spaced electrodes, and the magnetic sensors are positioned at the circumference of the bed, preferably on a plurality of planes along the vertical axis of the vessel;
- b) the plurality of electrodes comprise a pair of radially spaced electrodes, and the magnetic sensors are positioned at the circumference of the bed, preferably on a plurality of planes along the vertical axis of the vessel;
- c) the plurality of electrodes comprise a pair of axially spaced electrodes, and the magnetic sensors are arranged above and / or underneath the catalyst bed;
- d) the plurality of electrodes comprise a pair of radially spaced electrodes, and the magnetic sensors are arranged above and / or underneath the catalyst bed.

The terms "axially spaced" or "radially spaced" are understood as relative to the vertical axis of the vessel.

- A first set of magnetic sensors may be distributed at equally spaced angles across the azimuth in a first plane, and at least one further set of magnetic sensors may be distributed at equally spaced angles across the azimuth in a further plane along the axis of the reactor. The number of magnetic sensors in a plane may be from 2 to 36, in particular from 4 to 15, e.g., 6. The number of planes may be from 2 to 200, in particular from 3 to 10, e.g., 7. Fig. 2 shows a sensor arrangement of 8 magnetic field sensors in 7 planes.

- The quality of the reconstruction of the electric field depends on the radial distance of the sensors, the number of sensors per plane and the number of planes. The best sensor configuration may be determined for each reactor separately in an optimization process taking into account the desired resolution, i.e., the smallest size of agglomerates and void spaces to be measured.

- The distance between the sensors in azimuthal and vertical direction depend on the bed properties to be detected. For instance, the number of sensors might be increased in a certain location of the reactor, in order to obtain a more precisely resolution of the magnetic field, because the accurate position or size of inhomogeneities in this region is crucial for the operation of the process.
- 5 The minimum distance of the planes D_p is not particularly limited and the sensors may be mounted closely on top of each other; the maximum distance is half of the distance H of the electrodes. In order to avoid ambiguities in reconstructing the electric field from the magnetic field data, magnetic sensors should be provided in at least one plane at the location of the electrodes. Preferably, the distance is $D_p \approx H/2, H/3, \dots, H/200$ or smaller. The distance D_p has not to be constant for the whole reactor. A concentration of planes at
- 10 a specific region of interest is also preferable, e.g. the distance $D_{p, mid}$ in the middle of the reactor can be two times smaller than the distance at the upper and lower third of the reactor.

- The minimum radial distance between the outer diameter D_b of the electrical conducting bed and the sensor is 0, when the sensors are directly located at the outer surface of the conducting bed. Generally, the maximum radial distance is three times D_b , preferably, $D_b/4$ to D_b .
- 15 For example, if an inhomogeneity of the conductivity has the diameter D_v , the distance of the planes may be in the range $D_v/2 < D_p < 10 D_v$. Smaller and larger distances might be also possible. The detectable range of inhomogeneities is given $1 \leq D_v/D_b \leq 0.01$, preferably $1 \leq D_v/D_b \leq 0.1$.

- The magnetic sensors can be any of the various types of magnetic field sensors. All magnetic field components, e.g. tangential, radial and vertical components, may be measured and all components of the spatial and temporal derivatives of the magnetic field can also be measured and included in the evaluation.
- 20 The appropriate selection of the sensor positions and the magnetic field components to be measured is crucial in order to avoid ambiguities in the reconstruction of the electric field.

- An important feature of the measuring system is that the individual sensors are arranged in a predetermined array that is fixed in relation to each other and with respect to the reactor.
- 25 With the preferred electric current density, the strength of the magnetic field is generally from 0.1 to 1.200.000 nT, preferably from 5 to 100.000 nT. Higher magnetic fields are also possible, e.g. 1 to 100 mT.

- The magnetic sensors can be any magnetic field sensing elements and may be selected from, but are not limited to fluxgate probes, Hall probes, planar Hall probes, induction coils or combinations thereof. Preferably, the resolution of the magnetic sensors is from 0.1 nT to 1.2 mT. The magnetic sensors may
- 30 comprise gradiometric probes, which are able to determine the spatial derivative of the magnetic field in various orientations.

- Fluxgate probes (fluxgate sensors) have an exciter coil, a ferromagnetic core and a read-out coil. They are suitable for the measurement of weak magnetic fields as the magnetic field to be measured is chopped with the aid of the exciter coil and the ferromagnetic core, i.e., is periodically switched on and off at the location
- 35 of the read-out coil.

- A Hall probe (Hall generator plate) is a conductor or a semiconductor, the resistance of which varies as a function of the product of a magnetic field applied at right angles to the element and the current flowing in

the element. When a Hall probe is used, an output voltage from such a device results from the Lorentz force of the magnetic field acting on charge carriers within such a device.

5 The magnetic sensor can also be based on the planar Hall effect of ferromagnetic materials. The measurement principle is based on the change in anisotropic magnetoresistance caused by an external magnetic field in the Hall geometry. In contrast to a normal Hall sensor, which can in particular detect the field components perpendicular to the sensor plane, the planar Hall sensor has a high sensitivity with regard to the magnetic field components in the sensor plane. An asymmetrical electric field is measured perpendicular to the current, which depends on the magnetization state of the sensor.

10 Several magnetic sensors can be combined to give a gradiometer. Such modified sensors are able to measure spatial gradients of the magnetic field.

When performing a chemical reaction in a particle bed or in a regular structure having a plurality of parallel channels, unwanted depositions on the particle surface or the monolith walls may occur. In many cases, these depositions form a continuous electrically conductive layer. This layer causes a disturbance in the electric field, which may be detected by the method according to the invention.

15 If, in the course of a process, chemical reactions cause the proportion of electrically conductive particles or the value of the electrical conductivity of matter in partial volumes of the reactor to change, these changes can be recorded using the inductive measuring system. After appropriate evaluation and data analysis, this data can be used to localize regions of high chemical reaction activity, to characterize the reaction dynamics or to detect deviations from the specified process sequence.

20 In a particle bed, the spatial distribution of current density can be used to analyze irregularities in particle distribution such as agglomerates or voids, as well as particle motion. Agglomerates have higher electrical conductivity than the bulk of loose, non-agglomerated particles. As a result, they lead to a constriction of the field lines, which is detected via the magnetic field. The size of the agglomerates correlates with the strength of the disturbance in the magnetic field. Voids or cavities can be detected in an analogous way where electric
25 field lines tend to be much more dense near the perimeter of a void in the particle bed. Via the spatial displacement of disturbances in the moving bed, the migration velocity of the bed can be determined by tracking the migration of the signal patterns that are associated with the respective disturbances.

Any increase of the electric field density causes the volumetric heat flux to increase proportionally. In particular in a reactor with a large cross-sectional area, spatial inhomogeneity of the electric current
30 distribution and thus of the heat release leads to an appreciable non-uniform temperature distribution over the cross-section.

An initial imbalanced distribution of particles in the bed may be random or the result of asymmetries. Without countermeasures, such imbalances can be self-amplifying. Higher current flow increases resistive heat release, which causes the temperature to rise. At higher temperature, the reaction rate increases. In
35 reactions that proceed with carbon deposition, e.g., hydrocarbon pyrolysis, high reaction rate accelerates carbon deposition. This in turn lowers the electrical resistance, so that even more current flows in this region.

A non-uniform temperature distribution may also be problematic because in kinetically limited processes, optimal yield is only possible at the optimal temperature. In areas of too low temperature, conversion is usually too low, and at too high temperatures, undesirable side reactions can occur or a catalyst can be

deactivated. In endothermic reactions, self-amplification can also be caused by catalyst deactivation, e.g., the catalyst deactivates fastest in the warmest regions, so that the chemical reaction dampens the resistive heat release less and the temperature rises further.

5 Appropriate countermeasures can be taken when specific thresholds are reached. Such measures may include gasification of the deposits, adjustments of the operation parameters for targeted removal of unwanted depositions that are likely to form agglomerates (e.g. enhancement of solid mass flow) or adjustments in order to reduce the further formation of such layers.

10 The plurality of magnetic field measurements are suitably processed to establish (reconstruct) the generating electric field. Establishing electric field characteristics may include applying an inverse algorithm preferably involving a regularization scheme. The inverse algorithm may be coupled with machine learning methods.

15 Starting from a network of magnetic sensors positioned around the reactor, whose position is known, the structure of the surrounding magnetic field is measured. From the measurement data received from the magnetic sensors, the generating electric field is calculated using an inverse algorithm with the help of a processor. With the knowledge of the spatial structure of the electric field, i.e. the electric current distribution in the reactor, inhomogeneities can be reported on a qualitative basis or spatially resolved.

In an embodiment, the locations of the magnetic sensors outside the bed may be determined by an iteration scheme of forward simulations and hypothetical reconstructions to identify an optimal arrangement of the magnetic sensors in order to minimize ambiguities in establishing the electric field characteristics.

20 Establishing electric field characteristics preferably includes compensating in the measured magnetic field data contributions that emanate from sources other than the electric current flowing through the reactor bed.

Hence, the data processing preferably includes a compensation for external influences, in particular by the ambient temperature and changes of the geomagnetic field. Hence, the data processing preferably includes an ambient temperature measurement by a thermocouple and/or geomagnetic field measurement by a reference sensor.

25 In addition, the magnetic fields due to the power supply cable to and from the reactor must be taken into account and possible spatial changes of the power supply cable should be taken into account. The compensation preferably comprises providing at least one additional magnetic sensor for measuring all components of the magnetic field at a distance from the reactor, at which the magnetic field generated by the current flow in the bed has decayed below a predetermined threshold value which could be smaller than 1 nT, for example. Interfering magnetic fields may be measured with one or two 1D, 2D or 3D probes, preferable one or two 3D probes outside the reactor at a distance to the reactor, where the strength of the magnetic field generated by the current through the reactor has decayed sufficiently. Generally, the distance of the sensor to the reactor has to be at least 6 times the diameter D_b of the electrical conducting bed. Larger distances up to 40 times of D_b are also possible. These measurement data serve as input for the compensation algorithm. A possible compensation method is described by Sieger et al. "Challenges in Contactless Inductive Flow Tomography for Rayleigh-Bénard Convection Cells", *Magnetohydrodynamics* 58 (2022), pp. 25-32. The environmental magnetic field is subtracted from each sensor according to its measurement direction. The magnetic field sensors itself may require a dynamic range of 6 orders of

magnitude. The mounting of the sensors may be very stiff to avoid changes of the location of the sensor relative to the reactor vessel.

- In order to reconstruct the current distribution in the reactor from the magnetic field measurements outside the reactor, a mathematical model is required. The model can be a FEM (Finite Element Model), a BEM (Boundary Element Model) or an Integral Equation model. The essential part of the model is Biot-Savarts law, which calculates the magnetic field $\vec{b}$ at one given location $\vec{r}$ taking the entire volume V , in which the current $\vec{j}$ is flowing, into account:

$$\vec{b}(\vec{r}) = \frac{\mu_0}{4\pi} \iiint_V \frac{\vec{j}(\vec{r}') \times (\vec{r} - \vec{r}')}{|\vec{r} - \vec{r}'|^3} dV'$$

- For the inversion, it is essential to force that the current is divergence free ($\nabla \cdot \vec{j} = 0$). The inversion itself can be implemented as an optimisation problem, taking the operational parameters of the reactor into account. Methods include the direct modeling of the bed similar to the methods for CIFT and MIT or the use of AI by for example using invertible neural networks.

- The processor may be a custom computational and/or logic circuit that records the measured magnetic fields, compensates the measured magnetic field according to the temperature and the environmental magnetic field, solves the inverse problem in real-time and stores the reconstructed current distribution. It may also transfer relevant information to the controller of the reactor. It may include a central processing unit (CPU) coupled to a storage device and a memory. The CPU can be a general purpose processor, a custom-designed processor, a microprocessor, or any other type of processor or circuit that can execute software instructions.

- The plurality of magnetic field measurements and processing thereof provide near real-time information about the three-dimensional distribution of the electric current density.

Another embodiment of the invention resides in coupling a high-resolution real-time reconstruction of the electrical current distribution in the bed with a validated model of the process in the reactor in order to calculate a set of control variables with which the process can be controlled and/or optimized.

- The information thus gained from the processed magnetic field measurements may be used to adjust one or more manipulated variables of the unit operation. This may be affected by a continuous variation or switching of suitable manipulated variables using, e.g., an open-loop or closed-loop control circuit.

- Manipulated variables include reactant flow; reactant concentration in the fluid stream which is flowing through the bed; downward particle velocity; heat input of the heating system (e.g. direct electrical heating, inductive heating, trace heating); and the degree of preheating of the fluid stream.

In the case of several electrodes and counter-electrodes, applied voltages may be varied individually in order to counteract the uneven current distribution.

In an exemplary embodiment, once an agglomerate or plug in a moving particle bed has been identified by means of the processed magnetic field measurements, the supply of reactant gas can be interrupted or

switched to an inert gas to prevent the agglomerate or plug from growing, until the agglomerate or plug leaves the reactor.

The method of the invention may be applied to a wide variety of chemical reactions. The chemical reaction may simply involve the decomposition of a reactant, but may also involve other reactants, in particular other
5 gaseous reactants or solid reactants such as particles constituting the particle bed.

In preferred embodiments, the chemical conversion is selected from

- (i) reforming of ammonia according to the idealized equation
 $2 \text{NH}_3 \rightarrow \text{N}_2 + 3\text{H}_2$
over a bed of particles of transition metal (such as Fe or Ni) supported on a refractory support
10 material;
- (ii) decomposition of methanol according to the idealized equation
 $\text{CH}_3\text{OH} \rightarrow \text{CO} + 2 \text{H}_2$
over a bed of Cu catalyst;
- (iii) reforming of methanol according to the idealized equation
15 $\text{CH}_3\text{OH} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + 3 \text{H}_2$
over a bed of Cu catalyst;
- (iv) Reverse Water Gas Shift Reaction according to the idealized equation
 $\text{CO}_2 + \text{H}_2 \rightarrow \text{CO} + \text{H}_2\text{O}$
over a bed of Ni catalyst;
- 20 (v) Methane Steam Reforming according to the idealized equation
 $\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3 \text{H}_2$
over a bed of SiC, C, Ni catalyst or Fe catalyst;
- (vi) Methane Dry Reforming according to the idealized equation
 $\text{CH}_4 + \text{CO}_2 \rightarrow 2 \text{CO} + 2 \text{H}_2$
25 over a bed of SiC, C, Ni catalyst or Fe catalyst;
- (vii) formation of hydrocyanic acid by the reaction of ammonia with hydrocarbons according to the idealized equation
 $\text{CH}_4 + \text{NH}_3 \rightarrow \text{HCN} + 3 \text{H}_2$
over a bed of carbon particles;
- 30 (viii) formation of hydrocyanic acid by formamide cleavage according to the idealized equation
 $\text{HCONH}_2 \rightarrow \text{HCN} + \text{H}_2\text{O}$
over a bed of stain less particles or Fe catalyst;
- (ix) Boudouard reaction according to the idealized equation
 $\text{CO}_2 + \text{C} \rightarrow 2 \text{CO}$
35 over a bed of carbon particles;
- (x) Methane Pyrolysis according to the idealized equation
 $\text{CH}_4 \rightarrow \text{C} + 2 \text{H}_2$
over a bed of carbon particles;

- (xi) Cracking of hydrocarbons according to the idealized equation

$$C_{(n+m)}H_{(2n+2m+2)} \rightarrow C_nH_{(2n)} + C_mH_{(2m)} + H_2$$
over a bed of carbon;
- 5 (xii) dehydroaromatization of methane according to the idealized equation

$$6 CH_4 \rightarrow C_6H_6 + 9 H_2$$
over a bed of Mo catalyst, optionally a mixed bed of Mo catalyst and carbon particles;
- (xiii) alkane dehydrogenation according to the idealized equation

$$C_nH_{(2n+2)} \rightarrow C_nH_{(2n)} + H_2, \text{ wherein } n = 2, 3, 4$$
over a bed of FeO/Pt catalyst, optionally over a mixed bed of FeO/Pt catalyst and carbon particles;
- 10 (xiv) styrene synthesis according to the idealized equation

$$C_8H_{10} \rightarrow C_8H_8 + H_2$$
over a bed of FeO/Pt catalyst, optionally over a mixed bed of FeO/Pt catalyst and carbon particles;
- (xv) formation of anhydrous formaldehyde according to the idealized equation

$$CH_3OH \rightarrow CH_2O + H_2$$
15 over a bed of Ag catalyst;
- (xvi) cyclohexane dehydrogenation according to the idealized equation

$$C_6H_{12} \rightarrow C_6H_6 + 3 H_2$$
over a bed of Pt catalyst;
- (xvii) alcohol dehydration according to the idealized equation

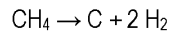
$$C_nH_{(2n+1)}OH \rightarrow C_nH_{(2n)} + H_2O, \text{ wherein } n = 2, 3, 4$$
20 over a bed of zeolite catalyst, optionally including carbon particles;
- (xviii) vinyl formamide synthesis from cyanoethyl formamide according to the idealized equation

$$CH_3CH(CN)(NH-COH) \rightarrow CH_2CH(NH-COH) + HCN$$
over a mixed bed of potassium hydroxide on alumina and carbon particles, or carbonized catalyst
25 of potassium hydroxide on alumina;
- (xix) melamine synthesis according to the idealized equation

$$6 (NH_2)_2CO \rightarrow C_3N_6H_6 + 6 NH_3 + 3 CO_2$$
over a mixed bed of bauxite and carbon particles, or carbonized bauxite;
- 30 (xx) oxidation of sulfur dioxide to sulfur trioxide as first step in the production of sulfuric acid, according to equation

$$2SO_2 + O_2 \rightarrow 2SO_3$$
over a bed of vanadium(V) oxide catalyst;
- (xxi) calcination of catalysts by decomposition of, e.g., nitrates, carbonates and hydroxides;
- 35 (xxii) drying of granulates in agricultural, wood and food industries; in paper, animal feed and pellet production; or in cement and recycling industry;
- (xxiii) regeneration of adsorbents, e.g., activated carbon; and
- (xxiv) regeneration of carbonized catalysts, e.g., desulfurization catalysts like Mo-Co on gamma-alumina, MTO catalysts and styrene catalysts.
- 40 Preferably, the chemical conversion is selected from (x) and (xi).

A particularly preferred embodiment of the invention relates to a hydrocarbon pyrolysis process (the conventionally used term methane pyrolysis will be used in the following and is intended to be synonymous with "hydrocarbon pyrolysis process"). Methane pyrolysis is a technology that splits hydrocarbons, especially aliphatic hydrocarbons such as methane, ethane, propane and/or butane directly into hydrogen and solid carbon. Most often, natural gas comprising methane is used as feedstock for methane pyrolysis. For methane, the pyrolysis proceeds according to the following main reaction:



The process is moderately endothermic (standard reaction enthalpy: 74.91 kJ/mol of CH₄). It is evident from the reaction equation above that in methane pyrolysis, the release of greenhouse gases is prevented. Therefore, in the event that the electric energy originates from renewable resources, methane pyrolysis is a CO₂-free, i.e., clean technology to obtain emission-free hydrogen. Methane pyrolysis is a one-step process which produces hydrogen in high volume.

As the energy demand of the methane pyrolysis contributes to the energy-penalty of the carbon fixture, a methane pyrolysis process having an as high as possible energy efficiency is preferred. Moving carbon bed methane pyrolysis is preferred due to several aspects, such as higher hydrogen yields.

The hydrocarbon feedstock contains at least one hydrocarbon which may be selected from methane, saturated hydrocarbons, in particular saturated C₂ to C₃ hydrocarbons, or C₅₋₉ hydrocarbons. The hydrocarbon feedstock preferably includes methane as a main hydrocarbon constituent, e.g., at least 75 vol.-% of methane, preferably at least 85 vol.-% of methane. In an embodiment, the hydrocarbon feedstock is natural gas.

The obtained solid carbon can be sold as a commercial product for selected applications, depending on the carbon morphology and physical/chemical properties. For example, the solid carbon from methane pyrolysis may be used for aluminum and steel production, tire manufacturing, electrode manufacturing, polymer blending, additive for construction materials, carbon devices like heat exchangers, soil conditioning, or storage.

Carbon particles act as the electrically conductive particles and as substrate for the deposition of carbon generated by the decomposition of the hydrocarbon compounds. The carbon particles can either be porous or non-porous and can be a granular or powder-like material. The particle size of a preferred support substrate is in the range of 0.3 to 20 mm, preferably 0.5 to 15 mm, more preferably 1 to 10 mm. The pyrolysis is preferably carried out at temperatures ranging from 1000 to 2500 K and at pressures ranging from 0.5 to 10000 kPa (abs).

A preferred method comprises the steps of:

- α) pyrolysis of hydrocarbons over the fluidized and/or moving bed of carbon particulates whereby carbon is deposited on the surface of the carbon particulates;
- β) recovering a stream of hydrogen-containing gas;
- γ) directing the stream of hydrogen-containing gas to a gas separation unit to obtain pure hydrogen and hydrogen-depleted gas;
- δ) recirculating the hydrogen-depleted gas to the reactor;

- ε) withdrawing carbon particles at the bottom of the reactor; and optionally
 - ζ) classifying the withdrawn carbon particles, preferably via sieving;
 - η) discharging a fraction of withdrawn carbon particles as a product;
 - θ) recirculating a fraction of withdrawn carbon particles directly to the reactor, and optionally
- 5 i) treating and activating a fraction of withdrawn particles and recirculating the treated fraction to the reactor.

A stream of a hydrocarbon feedstock enters the reactor where it is thermocatalytically decomposed (pyrolyzed) over the fluidized and/or moving bed of the carbon particles.

- Carbon particles are fed to the reactor at the top of the particle bed, and slide downwardly under gravitational field as a continuous column. The carbon particles are withdrawn at the bottom of the reactor. Subsequently, the withdrawn carbon particles may be classified, i.e., sorted by their particle size, into fractions. A (coarse) fraction of the withdrawn carbon particles is discharged as a product. A (middle-sized) fraction of the withdrawn carbon particles is fed directly back into the inlet. A (fine) fraction of the withdrawn carbon particles may be treated, e.g., ground into fine powder in a grinder and sieved, activated and/or formulated and recirculated to the reactor.

The gaseous residence time within the reaction zone is approximately 0.1 to approximately 600 s, preferably approximately 1 to approximately 60 s.

- A hydrogen-containing gas is withdrawn from the reactor. The hydrogen-containing raw product gas may pass a gas-solid separation unit. A filter, a cyclone, or any other system capable of separating fine particles from a gas stream, may be employed as the gas-solid separation unit. The purified gas stream is optionally directed through a heat exchanger and further to a gas separation unit. A gas separation membrane, a pressure swing adsorption (PSA) system, a cryogenic absorption (or adsorption) unit, or any other system capable of separating hydrogen from hydrocarbons, may be employed as the gas separation unit. A stream of pure hydrogen is separated from the gaseous stream.

- 25 A hydrogen-depleted gas is recirculated to the reactor.

The invention is illustrated by the accompanying drawings.

Fig. 1 is an illustration of the magnetic field induced by an electric current flowing through a longitudinal section of the reactor with a homogenous conductivity of the conducting bed.

- Fig. 2 is an illustration of an arbitrary sensor arrangement around a longitudinal electrically conducting bed using 56 magnetic field sensors.

Fig. 3 is an illustration of electric field lines in a homogeneous moving bed in a longitudinal section of the reactor (Fig. 3a), in a moving bed comprising a void space (Fig. 3b), and in a moving bed comprising an agglomerate (Fig. 3c).

- Fig. 4 shows the radial magnetic field component generated by the electrical current outside the conducting bed, as sensed by 24 magnetic field sensors located at different vertical locations, for a homogenous conductivity (Fig. 4a), a void space (Fig. 4b) and an agglomerate (Fig. 4c).
- Fig. 5 is a schematic view of a moving bed reactor adapted for carrying out the method according to the invention.
- Fig. 6 is a schematic view of a moving bed reactor adapted for carrying out the method according to the invention, wherein an electrical current flows radially.
- Fig. 7 is a schematic view of a fixed bed reactor adapted for carrying out the method according to the invention.
- Fig. 8 is a schematic view of a fluidized bed reactor adapted for carrying out the method according to the invention.
- While some of the embodiments below are illustrated with reference to an electrically conductive catalyst bed, it is understood that these embodiments may likewise be implemented with other electrically conductive beds.
- According to Fig. 1, the magnetic field induced by a current 102 in the conducting bed 101 with homogeneous conductivity has only an azimuthal component of the magnetic field visualized by the vectors 103 at several rings around the bed. The magnetic field curls around the conductor. The magnetic field can be illustrated by the right-hand rule. If one grasps the conductor with ones right hand, with ones thumb pointing in the direction of positive current flow 104, then ones fingers will curl around the conductor in the direction of the magnetic flux lines that encircle the conductor.
- Fig. 2 illustrates a magnetic sensor arrangement around the conducting bed 201 for the recording of the magnetic field of the electric current 202 in the conducting bed 201. The sensors 204 are arranged in 8 rings and 7 sensors per ring. This results in a total of 56 magnetic field sensors. Each sensor can either record all three components of the magnetic field or only selected ones, e.g. only the radial or vertical component of the magnetic field. In the situation illustrated in Fig. 2, a void space (white sphere) 203 is located in the conducting bed 201.
- Fig. 3 and 4 show the influence of the particle bed structure on the distribution of the electric current. A so-called forward simulation can be used to calculate how an assumed inhomogeneity modifies a given electric current distribution and its effect on the associated magnetic field. The simulation is usually based on an FEM model. Typical software packages are the open-source code Elmer, or Opera and Comsol. The set of parameters is based on operational data and equilibrium measurements for moving bed reactors using calcined petroleum coke (trade mark Carbon 99 from the manufacturer PINE-PACIFIC).
- The following reaction parameters underlay the simulation. In a cylindrical moving bed reactor with a diameter of 190 mm, a granulate of the calcined petroleum coke of the grain size fraction 2 mm to 4 mm is fed through the reactor from top to bottom. A gas stream with the feed composition H_2 : CH_4 = 75 vol.-% : 25 vol.-% is supplied from the bottom to the top. In the portion of the reactor shown, an electric current is conducted through the packing over a length of 1.4 meters. The current strength is 100 A and the current

density at the top and bottom edges is uniformly distributed over the cross-section. The granulate packing functions as an electrical heating resistor and generates temperatures of around 1250 °C.

The specific ohmic resistance of the particle bed depends on the loading of the carrier with pyrolysis carbon according to the equation:

$$\rho_{bed} = \rho_{\infty} + (\rho_0 - \rho_{\infty}) \cdot e^{-\frac{X_{PyC}}{\Delta X_C}}$$

wherein:

ρ_{bed}	specific ohmic resistance of the carbon bed		$\Omega \text{ m}$
ρ_0	specific ohmic resistance of the carrier	1.33e-02	$\Omega \text{ m}$
ρ_{∞}	specific ohmic resistance at high pyrolysis carbon loading (asymptotic limit)	7.18e-07	$\Omega \text{ m}$
X_{PyC}	pyrolysis carbon loading		kg_{PyC} / kg_{CPC}
ΔX_C	intrinsic pyrolysis carbon loading *	1.15e-02	kg_{PyC} / kg_{CPC}

* The intrinsic carbon loading is the loading of the granulate with pyrolysis carbon at which the tangent to the curve of the specific electrical resistance with unloaded bulk material intersects the asymptotic value of the specific electrical resistance at high pyrolysis carbon loading.

- 10 As methane pyrolysis takes place within the electrically heated zone of the reactor, the loading of the carrier with pyrolysis carbon increases in the axial direction from 0 to 2.5 mass-%. The diagram shows the electric current lines in a longitudinal section of the reactor.

According to Fig. 3, the electric field lines in a homogeneous moving bed in a longitudinal section of the reactor (Fig. 3a) with homogeneous conductivity are straight lines. In case a void space (white sphere) is located in the moving bed, the current has to pass around the sphere, because of low electrical conductivity (Fig. 3b). If an agglomerate with a tenfold higher electrical conductivity than of the bed is situated in the moving bed (dark sphere) (Fig. 3c), the current will be concentrated in the agglomerate.

Fig. 4 shows the radial component of the magnetic field at one azimuthal position for the same three cases of Fig. 3, as sensed by 24 magnetic field sensors located at different vertical locations. The strength and direction of the magnetic field is indicated by arrows at the sensor locations. If the conducting bed has a homogeneous conductivity, the radial magnetic field is zero (Fig. 4a). For both cases of a void space (Fig. 4b) or an agglomerate (Fig. 4c), the radial component of the magnetic field is non-zero and the two different cases can be clearly distinguished by the sign of the measured radial component. The distance between the conductive bed and the location of the magnetic field measurement is equal to the radius of the moving bed.

According to Fig. 5, a moving bed reactor has an electrically conductive catalyst bed 1 within a reactor shell 3. The reactor shell 3 may have a refractory lining (not shown in Fig. 5) between the catalyst bed 1 and the shell 3. Electrically conductive particles are fed via particle inlet 13 which are metered to the reactor, for

- example by a rotary valve (not shown in Fig. 5). The particles slide downwardly under gravity as the catalyst bed 1. The particles are withdrawn at the bottom of the reactor via particle outlet 14, for example controlled by a rotary valve (not shown in Fig. 5).
- 5 A voltage is applied across the catalyst bed 1 via electrodes 4 so that an electric current is induced to flow through the catalyst bed 1 between electrodes 4 to result in resistive heating of the catalyst bed 1. Electrical insulation 5 insulates the feedthroughs of the electrodes 4 from the reactor shell 3, i.e. the middle section of the reactor shell 3.
- 10 A fluid stream, which is typically a gaseous stream, is introduced via fluid inlet 11 and flowing through the catalyst bed 1. The fluid stream carries reactants, e.g. gaseous reactants, over the catalyst bed 1 and/or carries reaction products, e.g., gaseous reaction products, out of the catalyst bed 1. Fluid is withdrawn from the reactor via fluid outlet 12.
- Magnetic field sensors 6 are arranged at the outside of the reactor shell 3 to sense a magnetic field induced by the electric current flowing through the catalyst bed 1.
- 15 The electrical current fed into the catalyst bed may provide resistive heating to endothermic reactions. Via the magnetic field measurements, stagnation zones and agglomerates may be detected.
- Suitable endothermic reactions include methane pyrolysis on a bed of carbonaceous particles, hydrocarbon cracking to olefins on a bed of carbonaceous particles, and HCN synthesis using the Shawinigan process on a bed of carbonaceous particles.
- 20 According to Fig. 6, a moving bed reactor has an electrically conductive catalyst bed 1 in a zone having an annular cross-section within a reactor shell 3. Electrically conductive particles are guided via particle inlet 13. The particles slide downwardly under gravity as the catalyst bed 1. The particles are withdrawn at the bottom of the reactor via particle outlet 14.
- 25 A voltage is applied across the catalyst bed 1 via a pair of concentric cylindrical electrodes 4 so that an electric current is induced to flow through the catalyst bed 1 in a radial direction between electrodes 4 to result in resistive heating of the catalyst bed 1. Electrical insulation 5 insulates the feedthroughs of the electrodes 4 from the reactor shell 3.
- 30 A fluid stream, which is typically a gaseous stream, is introduced via fluid inlet 11 and flows in radial direction through the catalyst bed 1. The fluid stream carries reactants, e.g. gaseous reactants, over the catalyst bed 1 and/or carries reaction products, e.g., gaseous reaction products, out of the catalyst bed 1. Fluid is withdrawn from the reactor via fluid outlet 12. The fluid flow within the reactor is guided by the displacer element 7.
- Magnetic field sensors 6 are arranged at the outside of the reactor shell 3 to sense a magnetic field induced by the electric current flowing through the catalyst bed 1.
- 35 The electrical current fed into the catalyst bed provides resistive heating and induces a magnetic field for the detection of coking and agglomerate formation.

Suitable endothermic reactions include C_3H_6 synthesis using the UOP Oleflex process on Pt catalysts. A modified embodiment of the reactor according to Fig. 6 having a fixed catalyst bed is envisaged for the C_8H_8 synthesis on KFeO catalysts.

According to Fig. 7, a fixed bed reactor has an electrically conductive catalyst bed 1 within a reactor shell 3.

- 5 A voltage is applied across the catalyst bed 1 via electrodes 4 so that an electric current is induced to flow through the catalyst bed 1 to result in resistive heating of the catalyst bed 1. Electrical insulation 5 insulates the feedthroughs of the electrodes 4 from the reactor shell 3.

- A fluid stream, which is typically a gaseous stream, is introduced via fluid inlet 11 and flowing through the catalyst bed 1. The fluid stream carries reactants, e.g. gaseous reactants, over the catalyst bed 1 and/or
10 carries reaction products, e.g., gaseous reaction products, out of the catalyst bed 1. Fluid is withdrawn from the reactor via fluid outlet 12.

Magnetic field sensors 6 are arranged above and / or underneath the catalyst bed 1 within the reactor shell 3 to sense a magnetic field induced by the electric current flowing through the catalyst bed 1.

- The reactor according to Fig. 7 may be envisaged for carrying out exothermic reactions. The electrical
15 current fed into the catalyst bed provides excitation for magnetic induction tomography, preheating and ignition of the reaction.

Suitable exothermic reactions include NO synthesis according to the Ostwald process on Pt/Rh meshes, HCN synthesis in the Andrussov process on Pt/Rh meshes, H_2CO synthesis using the BASF process on Ag granulate.

- 20 According to Fig. 8, a fluidized bed reactor has an electrically conductive fluidized catalyst bed 1 within a reactor shell 3. Electrically conductive particles are fluidized by a gaseous stream introduced via fluid inlet 11 and are flowing through the catalyst bed 1.

- A voltage is applied across the catalyst bed 1 via electrodes 4, comprising a central electrode extending along the axis of the reactor and a generally cylindric counter electrode. Electrical insulation 5 insulates the
25 feedthroughs of the electrodes 4 from the reactor shell 3.

Magnetic field sensors 6 are arranged above the catalyst bed 1 within the reactor shell 3 to sense a magnetic field induced by the electric current flowing through the catalyst bed 1.

- Gaseous fluid is withdrawn from the reactor via fluid outlet 12. As shown in Fig. 8, the withdrawn gaseous fluid may be guided to a dust collector 7. In this case, a purified gaseous fluid obtained in dust collector 7 is
30 withdrawn via fluid outlet 12'. Solid particles are partially recycled via line 13 and partially withdrawn via line 14.

The electrical current fed into the catalyst bed may provide resistive heating to endothermic reactions. Via the magnetic field measurements, stagnation zones and agglomerates may be detected.

- Suitable endothermic reactions include methane pyrolysis on a bed of carbonaceous particles, HCN
35 synthesis using the Shawinigan process on a bed of carbonaceous particles and benzene synthesis by dehydroaromatization of methane on a bed of Mo_2C particles.

Claims

1. A method for evaluating and/or controlling a process in a vessel for unit operations involving an electrically conductive bed, comprising
 - 5 a) applying a voltage across at least a portion of the bed through a plurality of electrodes that are in an electrically conductive relationship with the bed to induce an electric current flowing through the bed; and
 - b) measuring a magnetic field and/or measuring changes in a magnetic field induced by the electric current by magnetic sensors at a plurality of locations outside the bed to generate magnetic field data.
- 10 2. The method according to claim 1, additionally comprising
 - c) establishing electric field characteristics, preferably including the topology and/or the dynamics of the electric field in the bed, using the magnetic field data,
 - d) calculating a distribution of matter in the bed and/or distribution of electrical conductivity in the bed using the electric field characteristics established in c).
- 15 3. The method according to claim 2, additionally comprising
 - e) adjusting one or more manipulated variables of the unit operation according to the distribution of matter in the bed and/or distribution of electrical conductivity in the bed obtained in d).
- 20 4. The method according to claim 2 or 3, wherein establishing electric field characteristics includes compensating in the measured magnetic field data contributions that emanate from sources other than the electric current flowing through the electrically conductive bed, wherein compensating preferably comprises providing at least one additional magnetic sensor for measuring all components of the magnetic field at a distance from the vessel, at which the magnetic field strength generated by the current flow in the bed has decayed below a predetermined threshold value.
- 25 5. The method according to any one of claims 2 to 4, wherein establishing electric field characteristics includes applying an inverse algorithm, preferably an inverse algorithm involving a regularization scheme, wherein the inverse algorithm is optionally coupled with machine learning methods.
- 30 6. The method according to any one of the preceding claims, wherein the locations of the sensors outside the bed are determined by an iteration scheme of forward simulations and hypothetical reconstructions to identify an optimal arrangement of the magnetic sensors in order to minimize ambiguities in establishing the electric field characteristics.
7. The method according to any one of the preceding claims, wherein the bed comprises a regular packing and/or a particle bed.

8. The method according to claim 7, wherein the particle bed is a moving particle bed, the method preferably comprising
feeding the particles to the vessel at the top of the particle bed,
allowing the particles to flow downwardly under gravitational field as a continuous column, and
5 withdrawing the particles from the vessel at the bottom of the particle bed.
9. The method according to claim 7, wherein the particle bed is a fluidized particle bed, wherein the particles are fluidized by an upwardly flowing gas injected at the bottom of the vessel to form a fluidized bed of particles.
10. The method according to claim 7, wherein the bed is a fixed particle bed.
- 10 11. The method according to any one of the preceding claims, wherein the electric current concurrently supplies electric resistance heating to the unit operation.
12. The method according to any one of claims 7 to 11, wherein the particle bed comprises electrically conductive particles which are preferably selected from carbonaceous particles, silicon carbide particles, metallic particles or composites thereof, and wherein the particle bed optionally contains
15 chemically active particles.
13. The method according to any one of the preceding claims, wherein the magnetic sensors are selected from fluxgate probes, Hall probes, planar Hall probes, induction coils or combinations thereof; wherein the magnetic sensors may comprise gradiometric probes, which are able to determine the spatial derivative of the magnetic field in various orientations.
- 20 14. The method according to any one of the preceding claims, wherein the electrodes and magnetic sensors have one of the following configurations:
- a) the plurality of electrodes comprise a pair of axially spaced electrodes, and the magnetic sensors are positioned at the circumference of the bed;
- 25 b) the plurality of electrodes comprise a pair of radially spaced electrodes, and the magnetic sensors are positioned at the circumference of the bed;
- c) the plurality of electrodes comprise a pair of axially spaced electrodes, and the magnetic sensors are arranged above and / or underneath the catalyst bed;
- d) the plurality of electrodes comprise a pair of radially spaced electrodes, and the magnetic sensors are arranged above and / or underneath the catalyst bed.
- 30 15. The method according to any one of the preceding claims, wherein the unit operation is selected from chemical conversions; and physical transformations such as drying operations; in particular from
- (i) reforming of ammonia according to the idealized equation
$$2 \text{ NH}_3 \rightarrow \text{N}_2 + 3 \text{ H}_2$$

- over a bed of particles of transition metal (such as Fe or Ni) supported on a refractory support material;
- (ii) decomposition of methanol according to the idealized equation

$$\text{CH}_3\text{OH} \rightarrow \text{CO} + 2 \text{H}_2$$
5 over a bed of Cu catalyst;
- (iii) reforming of methanol according to the idealized equation

$$\text{CH}_3\text{OH} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + 3 \text{H}_2$$
over a bed of Cu catalyst;
- (iv) Reverse Water Gas Shift Reaction according to the idealized equation

$$\text{CO}_2 + \text{H}_2 \rightarrow \text{CO} + \text{H}_2\text{O}$$
10 over a bed of Ni catalyst;
- (v) Methane Steam Reforming according to the idealized equation

$$\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3 \text{H}_2$$
over a bed of SiC, C, Ni catalyst or Fe catalyst;
- (vi) Methane Dry Reforming according to the idealized equation

$$\text{CH}_4 + \text{CO}_2 \rightarrow 2 \text{CO} + 2 \text{H}_2$$
15 over a bed of SiC, C, Ni catalyst or Fe catalyst;
- (vii) formation of hydrocyanic acid by the reaction of ammonia with hydrocarbons according to the idealized equation

$$\text{CH}_4 + \text{NH}_3 \rightarrow \text{HCN} + 3 \text{H}_2$$
20 over a bed of carbon particles;
- (viii) formation of hydrocyanic acid by formamide cleavage according to the idealized equation

$$\text{HCONH}_2 \rightarrow \text{HCN} + \text{H}_2\text{O}$$
over a bed of stainless particles or Fe catalyst;
- (ix) Boudouard reaction according to the idealized equation

$$\text{CO}_2 + \text{C} \rightarrow 2 \text{CO}$$
25 over a bed of carbon particles;
- (x) Methane Pyrolysis according to the idealized equation

$$\text{CH}_4 \rightarrow \text{C} + 2 \text{H}_2$$
30 over a bed of carbon particles;
- (xi) cracking of hydrocarbons according to the idealized equation

$$\text{C}_{(n+m)}\text{H}_{(2n+2m+2)} \rightarrow \text{C}_n\text{H}_{(2n)} + \text{C}_m\text{H}_{(2m)} + \text{H}_2$$
over a bed of carbon;
- (xii) alkane dehydrogenation according to the idealized equation

$$\text{C}_n\text{H}_{(2n+2)} \rightarrow \text{C}_n\text{H}_{(2n)} + \text{H}_2, \text{ wherein } n = 2, 3, 4$$
35 over a bed of FeO/Pt catalyst, optionally over a mixed bed of FeO/Pt catalyst and carbon particles;
- (xiii) dehydroaromatization of methane according to the idealized equation

$$6 \text{CH}_4 \rightarrow \text{C}_6\text{H}_6 + 9 \text{H}_2$$
40 over a bed of Mo catalyst, optionally a mixed bed of Mo catalyst and carbon particles;

- (xiv) styrene synthesis according to the idealized equation

$$\text{C}_8\text{H}_{10} \rightarrow \text{C}_8\text{H}_8 + \text{H}_2$$
over a bed of FeO/Pt catalyst, optionally over a mixed bed of FeO/Pt catalyst and carbon particles;
- 5 (xv) formation of anhydrous formaldehyde according to the idealized equation

$$\text{CH}_3\text{OH} \rightarrow \text{CH}_2\text{O} + \text{H}_2$$
over a bed of Ag catalyst;
- (xvi) cyclohexane dehydrogenation according to the idealized equation

$$\text{C}_6\text{H}_{12} \rightarrow \text{C}_6\text{H}_6 + 3 \text{H}_2$$
10 over a bed of Pt catalyst;
- (xvii) alcohol dehydration according to the idealized equation

$$\text{C}_n\text{H}_{(2n+1)}\text{OH} \rightarrow \text{C}_n\text{H}_{(2n)} + \text{H}_2\text{O}, \text{ wherein } n = 2, 3, 4$$
over a bed of zeolite catalyst, optionally including carbon particles;
- 15 (xviii) vinyl formamide synthesis from cyanoethyl formamide according to the idealized equation

$$\text{CH}_3\text{CH}(\text{CN})(\text{NH-COH}) \rightarrow \text{CH}_2\text{CH}(\text{NH-COH}) + \text{HCN}$$
over a mixed bed of potassium hydroxide on alumina and carbon particles, or carbonized catalyst of potassium hydroxide on alumina;
- (xix) melamine synthesis according to the idealized equation

$$6 (\text{NH}_2)_2\text{CO} \rightarrow \text{C}_3\text{N}_6\text{H}_6 + 6 \text{NH}_3 + 3 \text{CO}_2$$
20 over a mixed bed of bauxite and carbon particles, or carbonized bauxite;
- (xx) oxidation of sulfur dioxide to sulfur trioxide as first step in the production of sulfuric acid, according to equation

$$2 \text{SO}_2 + \text{O}_2 \rightarrow 2 \text{SO}_3$$
over a bed of vanadium(V) oxide catalyst;
- 25 (xxi) calcination of catalysts by decomposition of thermally decomposable compounds;
- (xxii) drying of granulates in agricultural, wood and food industries; in paper, animal feed and pellet production; or in cement and recycling industry;
- (xxiii) regeneration of adsorbents; and
- (xxiv) regeneration of carbonized catalysts.