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Development of a Clean Fluidized Bed Reactor for Food-Grade CO_2 Production

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ABSTRACT

Combustion of natural gas in fluidized bed reactors can be considered as an economical way of producing energy and food-grade CO_2 largely needed in food industries. Among the fluidized bed reactors, turbulent fluidized beds exhibit several advantages over conventional combustion methods, and bubbling and circulating fluidized beds. Operation of the turbulent fluidized bed at industrial levels may bring many advantages to food industry and therefore, their commercialization could be of great industrial importance. In this study, a new generation of fluidized bed reactors is developed along with proper inert particles to discover new applications in food processing. In order to develop such new reactors, a fluidized bed reactor is modified by adding secondary injection port so that the natural gas is properly and safely injected to the bed for the heat generation and CO_2 production purposes. As compared with the conventional CO_2 production facilities, the CO_2 produced in this way, due to small amount of emissions, has a very low purification cost. Moreover, the new devices could be well operated at medium temperatures and this feature could be of prime importance in commercializing this promising technology for food processing application, i.e., beverage industries.

1. INTRODUCTION

Traditional CO_2 production facilities, using fossil fuels, coal, oil derivatives or natural gas are operating at very high temperatures. Such operations are not only very expensive in terms of the costs associated with fuel and purification of combustion products, but also the emission levels remain very high and unacceptable. Furthermore, the CO_2 emitted to the environment leads to global warming, which is now a major focus of environmental concern and legislation. Few attempts were reported in the literature where CO_2 recovery from combustion devices has been considered for industrial applications, but considering all the advantages, this could only solve the global warming issue while the two other issues as outlined remain unanswered.

A traditional CO_2 generation assembly consists of the fuel-oil fed steam boiler with the necessary instruments, control and accessories necessary for the production of steam and flue gas. In this device, the amount of excess air is rather high as compared with the modern combustors proposed in this study. In addition, due to very high operating temperatures, some undesirable gases, i.e., SO_x, NO_x, CO, are generated increasing the purification cost associated with food-grade CO_2. The CO_2 generated in this way could not be easily used for food processing applications, i.e., beverage industries, which still remain the mega-consumer of food-grade CO_2. Therefore, a proper technology should be developed in order to overcome the shortcomings associated with the traditional combustion devices having met new environmental regulations. An increased availability of natural gas and the existing state-of-the-art modern technologies could serve as a framework to develop different attractive and cost-effective technologies in food processing area.

Any development of the new CO_2 generation facility should be based on lowering the combustor temperatures by using rather clean fuels, i.e., natural gas. This certainly results to a cost reduction for new facilities. The amount of such reduction depends on the way the new reactor is designed and operated and also the choice of bed materials. One should also keep in mind the safety problems associated with natural gas combustion. Among the modern technologies, the fluidized bed technology is a main choice as background for developing this novel technology. Basically, all fluidized bed reactors are operated based on the same principles in which gas or air is fed upward through a distributor at the bottom of the bed of finely divided particles. As the gas flows upward, fluidization occurs as the particles begin to move in liquid-like behavior.

Various fluidization regimes are observed as the superficial gas velocity is increased ranging from the bubbling regime to the slugging regime (for small beds), to turbulent and to the fast fluidization. The turbulent fluidization regime, which is regarded as the highly expanded gas-solid system, exists between bubbling and fast fluidization with degrees of expansion, between 0.65 and 0.85. This type of fluidization was first observed by Zenz but Lanneau has been the first who reported the existence of turbulent fluidization [1]. Compared to bubbling fluidized beds, turbulent fluidized beds have more diffuse and unclear upper surfaces which then lead to higher gas solid efficiency.

Turbulent Fluidized Bed (TFB) seems to be an ideal reactor for fast exothermic reactions at high temperatures, offering the advantages of exceptionally high heat transfer, intimate gas and solid contact, excellent thermal uniformity and temperature control, high overall productivity, versatility, much lower capital cost, high combustion efficiency and relatively short mean residence time as compared to the conventional combustion systems for a given throughput. Their overall homogeneous behavior makes the gas-solid contact to become more efficient by further enhancing the overall conversion [2, 3]. Therefore, these reactors can provide an innovative method of converting natural gas, to electricity with higher net efficiency and to food-grade CO_2 with a very low NO_x emission. They could also reduce pollutant emission level which is critical in food processing, packaging and preservation in order to address the national and environmental concerns by providing more reasonable combustor size and lower pollutant levels.

Although TFB technology is becoming mature from these commercial applications, there are some significant uncertainties in predicting their performance in large-scale systems. In addition, no information can be found in the literature regarding TFB applications in food processing. Attempts were made by Foka, et al. [4] to carry out the catalytic combustion of natural gas at low temperatures in a TFB of around 450°C and due to low amount of emissions; the CO_2 generated in this process can be used in food processing. Although, the catalytic combustion may be considered an innovation in food processing applications for practitioners, the combined catalysts, operational, maintenance and capital costs associated with this reactor are not competitive. Moreover, since the premixed fuel-air mixture is injected to the reactor in this way, the throughput remains very low (less than 4% methane in air). These disadvantages show that these reactors are not suitable for food industry.

In order to make fluidized bed reactors applicable for food processing applications and to decrease the costs, the catalysts should be replaced by inert materials. Therefore, the medium-temperature combustion in fluidized bed reactors is needed in order to reduce the associated costs while promoting complete combustion. The temperature lowering to medium level (800°C to 1000°C) leads to the following advantages in operation:

- High safety level in the combustion of flammable mixture, i.e., methane-air mixture by adding a secondary fuel injection port to the fluidized bed reactor design for direct fuel injection to the bed to any desired concentration level;

- Enhanced resistance of the reactor to corrosion;
- Prevention of the reactor deformation by temperature-related stresses that occur during normal furnace operations using stainless steel material to build the reactor; and
- Less emissions level.

Therefore, the main objective of the present study is to acquire a more fundamental understanding of this new technology through an extensive experimental program. Specific objectives are as follows:

- To characterize a proper inert material suitable for this novel technology;
- To develop and test the novel design by adding a secondary injection port to the bed and experimentally characterizing the hydrodynamics of this port;
- To measure CO destruction and formation profiles within the bed and inside the reactor to make sure of the suitability of the produced CO_2 for food processing applications; and
- To characterize the temperature and CO profile around the sparger.

2. EXPERIMENTAL APPARATUS AND PROCEDURE

In this section, the apparatus used for experiments and also the experimental procedure are described in details.

2.1 Apparatus

Two experimental setups are used in this investigation in order to study the behavior of the inert particles exposed to combustion in a fixed bed (FB) and also a pilot plant TFB reactor.

FB Reactor

A schematic diagram of the FB reactor is shown in Fig. 1. The reactor consists of two concentric tubes of 600 m length with 13 mm inner diameter (ID) and 7 mm outer diameter (OD). Its walls are made of alumina in order to limit wall catalytic effects. Combustion takes place between the two concentric tubes where the inert particles are carefully placed. Heating is provided by a high temperature furnace with a single heated zone of 222 mm long. The premixed methane-air mixture (2% to 4% methane, ranging from 100 and 500 ml/min) was fed to the reactor. A gas chromatograph equipped with a thermal conductivity detector (TCD) was used to monitor the methane and CO_2 mole fractions at the reactor exit. Inert particles, i.e., sand and alumina, of various average particle sizes are used during the combustion tests. For all experiments, the inner tube and furnace temperatures were measured under steady-state conditions.

Pilot TFB Reactor

The pilot TFB reactor used in this study consists of a 200 mm ID and 2 m tall refractory-lined reactor as shown in Fig. 2. The reactor is divided into four zones: the combustion inlet zone (wind-box

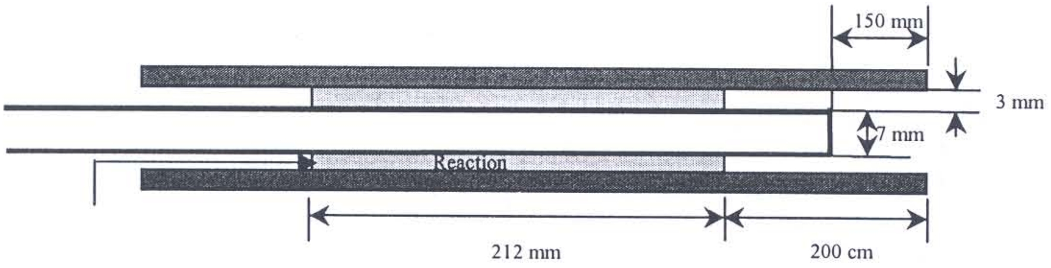
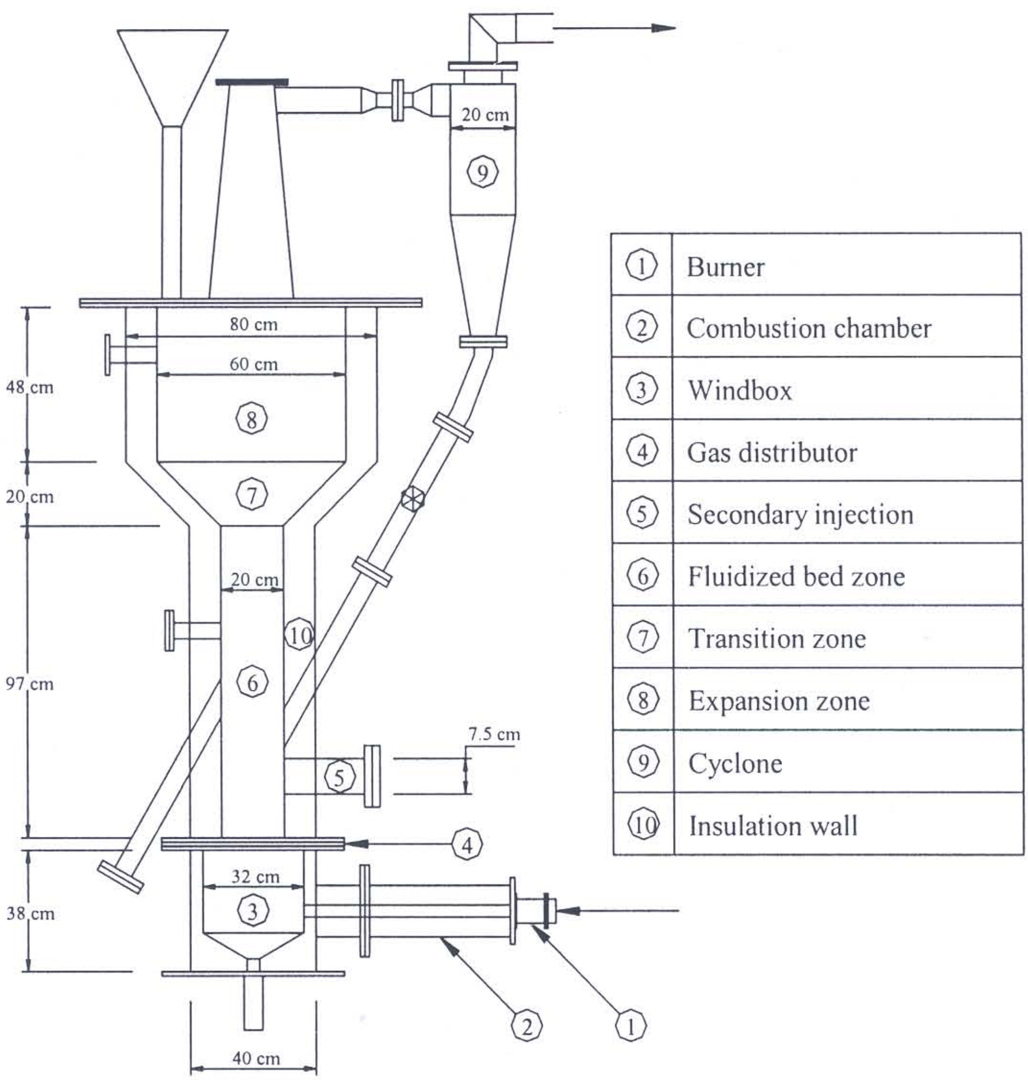


Fig. 1 Schematic diagram of the FB reactor



①	Burner
②	Combustion chamber
③	Windbox
④	Gas distributor
⑤	Secondary injection
⑥	Fluidized bed zone
⑦	Transition zone
⑧	Expansion zone
⑨	Cyclone
⑩	Insulation wall

Fig. 2 Schematic diagram of pilot TFB reactor

and distributor), a fluidized bed zone, a freeboard zone and an expansion zone of 600 mm ID. An external natural gas burner with the 20 kW nominal power located at the bottom of the bed provides the partial heat required for preheating the reactor to a desired temperature. Sand particles, tested in an FB reactor as described earlier, which consist mainly of silica with an average particle size of 543 mm are used during the combustion tests. Several ports are provided along the axial position for pressure measurements, sampling and natural gas injection to the reactor. Natural gas is supplied to the reactor through a secondary injection port with a sparger facing downward as shown in Fig. 3. The sparger of 6.33 mm in diameter was used throughout this study with one-hole of 4.33 mm in diameter. The sparger was kept free of particles during the preheating period using nitrogen as a purging fluid. Sampling probes were placed along the reactor centerline with their tips protected from particle clogging by a filter. The probes were connected to the gas chromatograph where the samples are withdrawn with a variable pressure vacuum system. Type-K thermocouples were also placed along the reactor centerline to monitor temperature profiles. An absolute pressure transducer was used to monitor the level of particles in the reactor by continuous recording of pressure fluctuations every second. Flow rates of air and natural gas were measured by orifice plates and rotameters, respectively. During the experiments, data are acquired by the data acquisition system where temperatures and pressures are recorded, respectively. At relatively high superficial velocities, a significant amount of particles are entrained. These particles are separated from the gas by a 0.2 m ID cyclone and re-circulated to the bed. An initial bed height of 2.5D (about 20 kg sand particles) was used in all experiments.

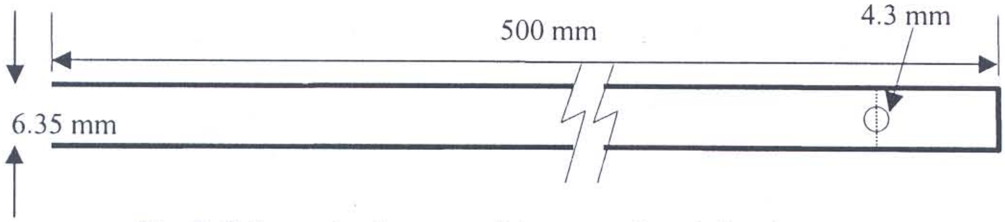


Fig. 3 Schematic diagram of the secondary injection sparger

2.1 Experimental Procedure

In this section, the details of the experimental procedure for fixed and fluidized bed studies are given.

FB Reactor

Fluidized bed reactors are hydrodynamically complex for deriving the kinetics information related to bed materials. Therefore, the data must be obtained in reactors (e.g., fixed beds) where hydrodynamics can be described confidently [5]. In this study, the emulsion phase of the fluidized bed reactors was approximated with a FB. This analogy can be justified considering the fact that the emulsion phase was usually assumed to be at minimum fluidization conditions, where the large amount of solid particles were present. Such analogy helped the authors to carry out the combustion of methane in a FB reactor to determine the effect of inert particles, i.e., sand and alumina on the combustion process. The details of the experimental procedure and modeling for methane combustion in a FB of inert particles are given elsewhere [6]. For the purpose of this study, different sizes of inert particles were used in the FB reactor and gas samples were drawn at the reactor outlet to analyze the combustion products. The reaction zone, which was a very thin shell of 3 mm, was well located at the reactor center. With a very small amount of mixture flowing through the reactor, the flow was fully developed and the reactor remained isothermal. Therefore, it was reasonable to assume the flow in the reactor as a plug flow.

Pilot TFB Reactor

Upon choosing the bed materials in the FB reactor, the experimental study in the TFB started by hydrodynamic tests, i.e., measuring minimum fluidization velocity, the onset of turbulent fluidization regime, etc. To carry out the combustion studies, the following procedure was adopted to heat up the insulated pilot TFB reactor to a desired temperature level:

- Raising the temperature up to 750°C, using the burner located at the reactor base;
- Raising of temperature up to 850°C, using the premixed combustion in wind-box, (under-bed combustion); and
- Raising the temperature up to 1000°C, using the non-premixed combustion, using the direct injection of NG to the reactor (over-bed and in-bed combustion).

It is important to note that in all experiments with the pilot plant, the solid inventory and temperature profile should be strictly controlled by means of pressure measurement and thermocouples, respectively. Attrition of particles in a fluidized bed reactor affects solid inventory of the reactor. At high gas injection velocities, highly turbulent area is created around the sparger. This may yield to generation of fine particles due to jet impingement. Under these conditions, the bed may easily become empty with excessive elutriation of resulted fine particles.

Upon reaching the bed temperature to the level of interest for combustion studies, natural gas is injected through the secondary injection port and the combustion takes place inside of the bed. One of the fundamental problems, which should be understood in developing any novel device for food processing applications, is to understand the CO formation and destruction pathway. In this way, one makes sure that its amount in the exit could easily be controlled. Since at the temperature ranges found in this study, the amount of NO_x formation is not significant and a considerable amount of information has been reported in the literature, less effort was put on this issue. Therefore, the local structure of the bed is characterized by measuring the concentration and temperature profiles around the sparger at the secondary injection port and also in the bed for various operating conditions. The typical experiment duration for TFB was 50 hours consisting of the reactor preparations, pre-heating, data acquisition, cooling down and data analysis.

3. RESULTS AND DISCUSSION

In this section, experimental data obtained in the FB reactor and the pilot plant fluidized bed reactor are discussed and compared with model predictions in some cases for a variety of operating conditions.

3.1 FB Reactor

Figure 4 shows the experimental data for empty reactor and for the FB reactor filled with sand and alumina particles. The figure also shows the plug flow model predictions [6] obtained with the Gas Research Institute methane combustion mechanism [7] for 2% initial methane mole fraction and mean residence time of 3 seconds. For the empty reactor, agreement comparison between experimental and predicted data is reasonably close to the inflection point, and the difference is attributed to the fact that

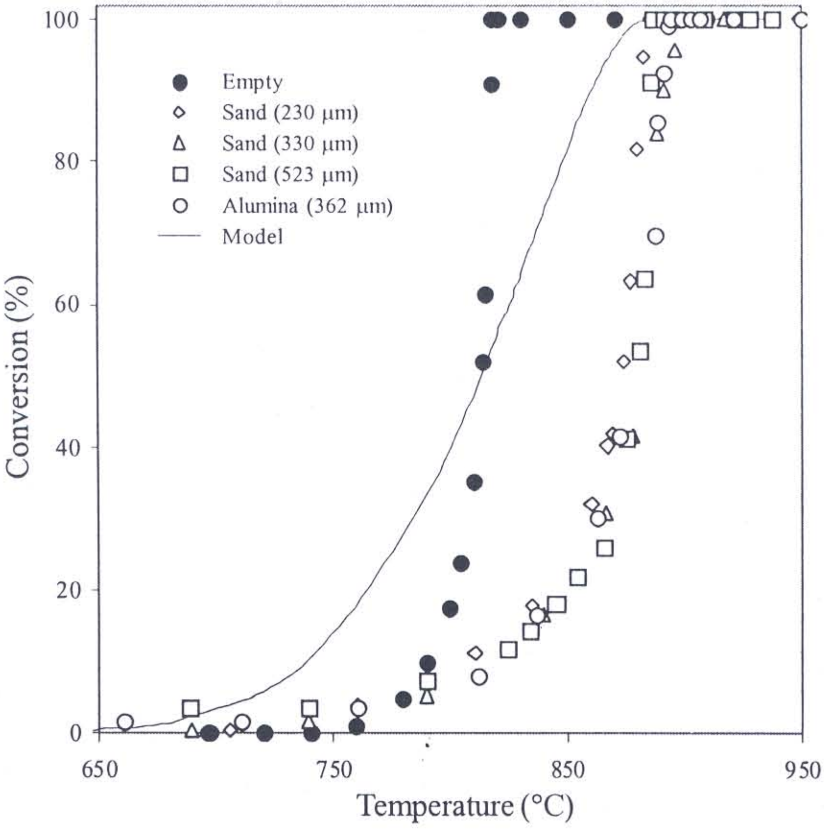


Fig. 4 Comparison of the experimental methane conversion data from an FB reactor with different sand and alumina particles and empty reactor to the predictions obtained from the model ($X_{CH40} = 2\%$, $\eta = 10^7$, $\tau = 3$ and $\tau_a = 2.4$)

the reactor walls may contribute to combustion. Alumina basically behaved like sand particles meaning that the reactor walls may also contribute to combustion. Experimental data and plug flow model predictions show the existence of two important temperature zones as follows:

- 700°C < T < ~875°C:

  At this zone, conversion at the empty reactor differs from the FB reactor. It seems that solid and wall surfaces alter homogeneous combustion by reducing free radical concentrations. Such inhibition effect may lower conversion as shown in Fig. 4. The same trend was reported for the premixed combustion in fluidized bed reactors [8, 9].
- T > ~875:

  In this zone, comparison between the model and experiment is satisfactory since the homogeneous combustion dominates and inhibition (heterogeneous reaction) becomes less pronounced. Hesketh and Davidson [10] showed that above a certain critical temperature, the emulsion phase exhibits homogeneous combustion behavior in a fluidized bed reactor. This would be a strong confirmation of the analogy made in this study between FB and TFB to choose the bed materials based on their contributions to the combustion reactions. Therefore, it is concluded that the inert particles tested in this study would be an excellent choice for bed materials used in fluidized bed studies to develop a novel reactor. Since the alumina and sand particles are behaving closely, economic consideration for industrial reactors suggests choosing of the sand particles as bed materials for TFB experiments.

In Fig. 5, a comparison between CO mole fraction calculated by mass balance and the plug flow model is presented. The CO mole fraction is low compared to model prediction for T < 875°C. This is a very interesting finding for food processing applications since it shows that at this temperature range, combustion over sand particles may lead to less CO emission (almost half) as compared with homogeneous combustion. This effect can be attributed to the low concentration of OH radicals as the main oxidant species of CO. The OH radicals are substantially reduced by recombination reactions at the particle surface. In addition, this figure shows that the consumption of CO is very small compared to its formation at lower temperatures leading to a maximum in CO concentration.

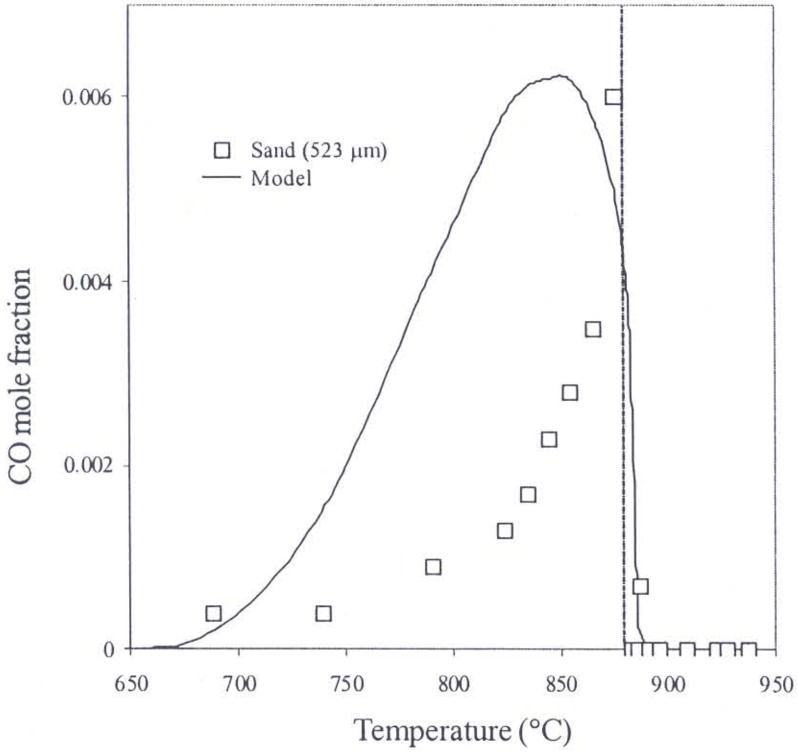


Fig. 5 Comparison of CO mole fraction from an FB reactor for the sand particles to the predictions obtained from the model (X_{CH40} = 2%, $\eta = 10^7$ and $\tau = 3$)

3.2 Pilot TFB Reactor

Figure 6 shows the normalized standard deviation of pressure fluctuation measured as a function of gas superficial velocity for sand particles at 920°C. The figure exhibits a well-defined maximum, which is interpreted as the onset of turbulent fluidization (U_c) based on the definition found in the literature [3]. It is important mentioning that most measurements of U_c have been done at ambient temperature. Its determination at high temperatures is very tedious since for every single point, the reactor must reach its steady-state conditions prior to pressure signal measurements. Based on the information generated in this figure, a typical superficial velocity of 1.5 m/s is chosen for the combustion experiments in turbulent fluidization conditions.

Figure 7 shows the axial bed temperature profiles for TFB during a typical experiment. Because of rapid solid mixing, TFB temperatures are quite uniform (variations within ±7°C). In fact, such temperature uniformity is of utmost importance in the development of the novel devices for food processing applications.

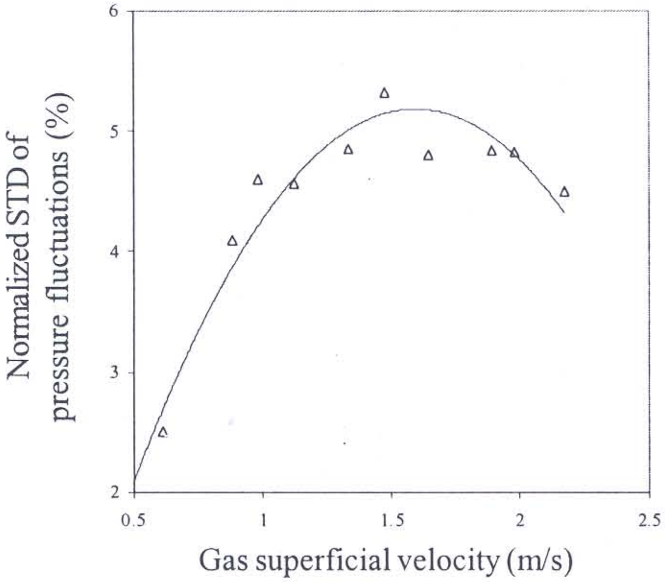


Fig. 6 Normalized standard deviation (STD) of pressure fluctuation at 920°C and ($d_p = 543$ μm, $z = 150$ mm and $D = 200$ mm) for onset of turbulent fluidization

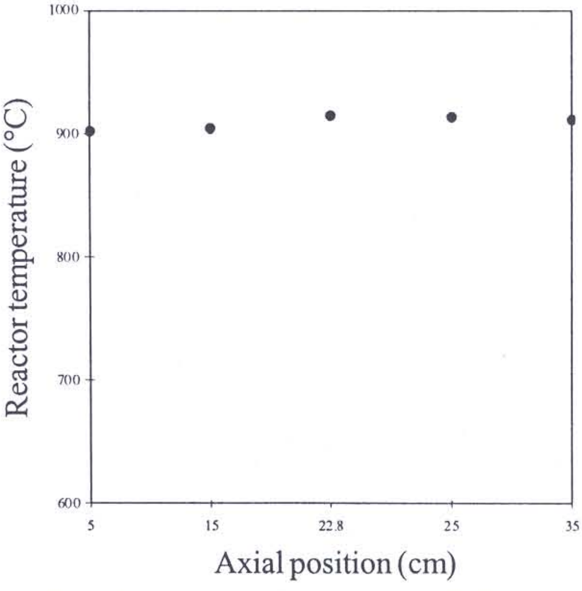


Fig. 7 Axial temperature profiles in turbulent flow regime with sparger placed at $z = 17$cm ($U_j = 120$ m/s)

A typical natural gas conversion profile obtained from the TFB experiments is shown in Fig. 8. For all experiments, the conversion is 100% at the top of the bed. As temperature increases, the 100% conversion point moves to the bed and this can be attributed to the fact that the free radial formation dominates the inhibition process.

In Fig. 9, CO measurements along the reactor height for TFB are reported. As seen in this figure, upon injecting the natural gas through the secondary injection port and due to the high turbulent area created around the sparger, excellent mixing was achieved between fuel and air. In this region, the amount of O_2 is much lower than the stoichiometric value needed to convert the fuel to CO_2 and almost all fuel was immediately converted to CO, leading to a peak in CO concentration and under this condition,

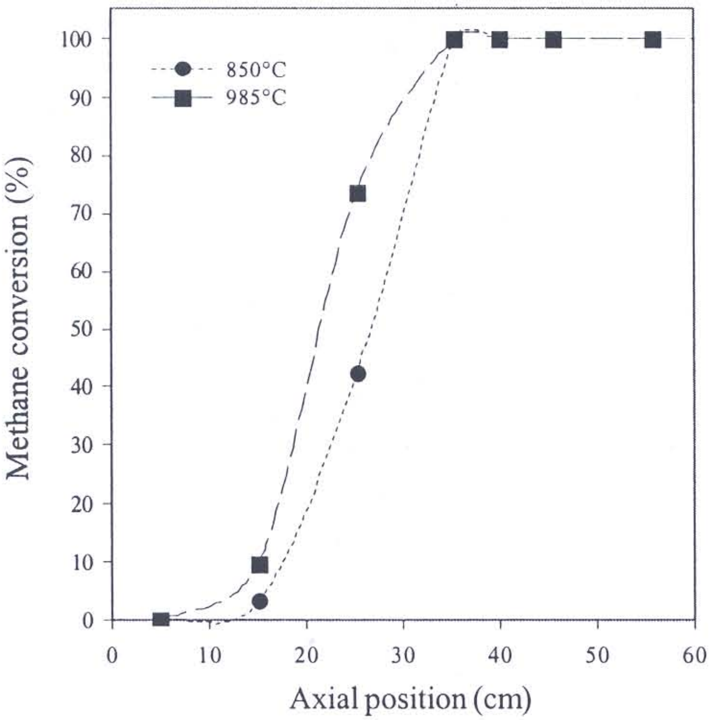


Fig. 8 Methane conversion profile in the turbulent regime at various temperatures with sparger placed at the reactor base

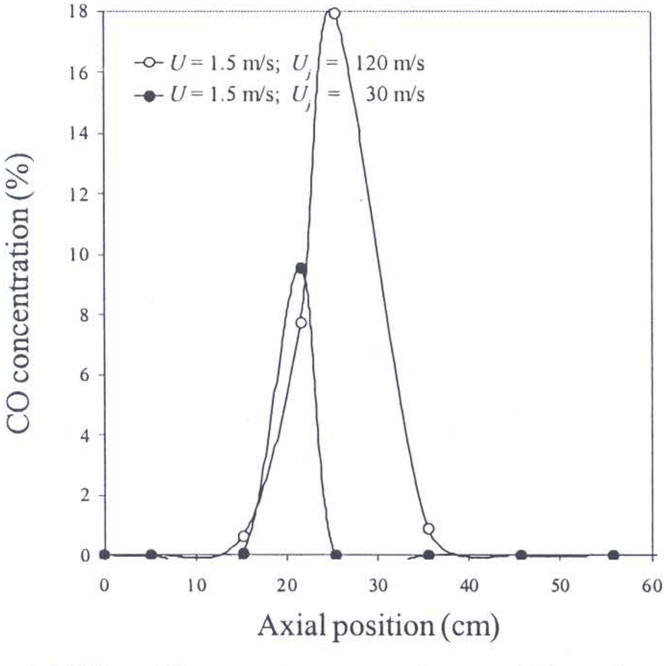


Fig. 9 Axial CO profile at various operating conditions for non-premixed combustion with one-hole sparger placed at $z = 17$ cm ($T \sim 900$°C)

CO-rich bubbles were formed. Therefore, it can be concluded from this figure that the fuel combustion took place in two consequent steps. Initially, the fuel was converted to CO around the jet and then CO was converted to CO_2, if the bed temperature was kept sufficiently high. The results of FB studies (Fig. 5) also confirm such findings. The complete CO oxidation in the turbulent fluidization can be achieved very fast as compared to bubbling fluidization (low gas superficial velocities). This can be attributed to the fact in TFB, the gas mixing is improved. For low bed temperatures, the observation of CO profile in the reactor leads to the conclusion that CO burns in the freeboard.

Figure 10 shows the concentration of CO measured at the TFB outlet. TFB generates less CO probably due to high gas-solid interactions as compared with the bubbling fluidized bed leading to low. This trend is also confirmed in the FB studies (Fig. 5). In CO_2 generation facilities, the amount of CO formation can go up to 1.2%. 0.4% CO which is very acceptable in these facilities, which is still considered very high as compared with the new technology. Such high CO level reduces the combustion efficiency. While in the novel reactor proposed in this study as shown in Fig. 9, the complete conversion of natural gas to CO_2 can be achieved. The measurement of CO formation inside of the reactor confirms this finding.

In Fig. 11, normalized NO_x concentration is reported for TFB along with predicted concentrations based on equilibrium calculations. It is important to note that the amount of NO_x generated for TFB conditions is always lower than the value measured with the burner located at the reactor base (40 ppm for stochiometric conditions). For industrial TFB reactors with integrated heat exchanger inside, due to low oxygen level, less NO_x may be measured as compared to those reported in this study. The amount reported for NO_x content in an empty chamber (flame combustion) is about 200 ppm where the temperature reaches to over 1700°C [11]. The NO_x formation in the fluidized bed reactor does not depend on the thermal load of the bed, while in traditional chambers; it grows up rapidly with the increase of the thermal load of the chamber.

3.3 Heat Generation

When the reactor was operated in the turbulent regime, 6% of methane was injected to the reactor and the conversion was 100% at the temperature range of interest. Superficial gas velocity was

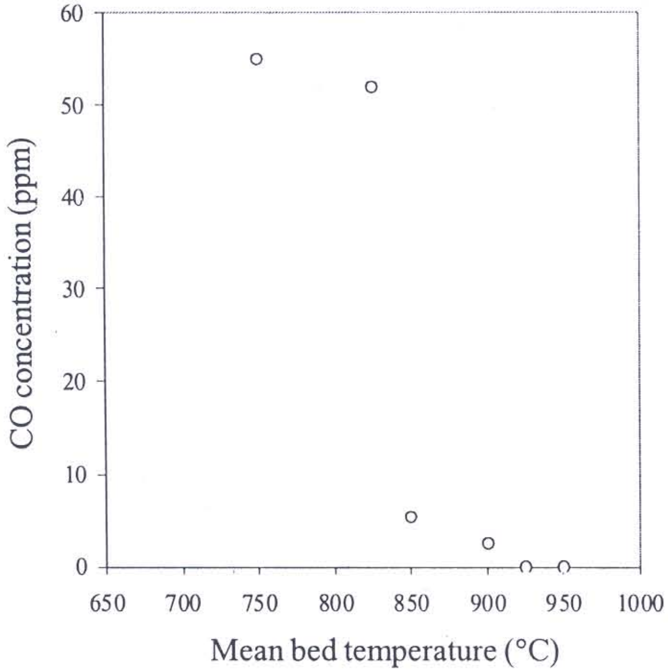


Fig. 10 CO emissions at reactor exit for turbulent regime with sparger placed at the reactor base

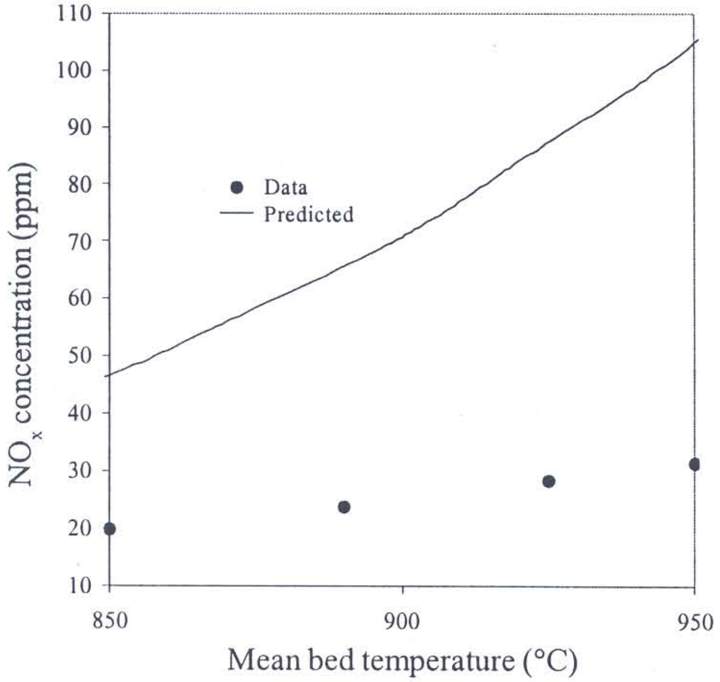


Fig. 11 Predicted and experimental NO$_x$ concentration at the reactor exit with sparger placed at the reactor base

1.5 m/s and a maximum temperature of 980°C was used. The reactor power generated in this investigation was about 42 kW for the turbulent regime. In CO$_2$ generation facilities, the energy could be used for adsorption process or in any other units found in food or beverage industry.

4. CONCLUSIONS

A clean reactor has been proposed and tested for food processing applications with proper inert bed materials using a TFB technology. The main results of this study as derived from the experimental observations are summarized as follows:

- Sand particles can be considered as an excellent and feasible choice as bed materials for the clean TFB.
- This study showed that combustion takes place at two steps and confirms the full destruction of CO inside of the bed. The results of measurements at reactor exits for both fixed and fluidized bed studies confirm the effect. For food processing applications, this is a very interesting result.
- Onset of turbulent fluidization was measured since the existing correlations cannot be used to determine this velocity for the condition range of this study.
- For TFB reactors, the complete natural gas conversion can be achieved at over 800°C.
- The power generated by TFB is much higher than that for bubbling fluidized bed reactors while respecting all environmental requirements.
- Measured CO profile showed that a reducing zone similar to the lower region of circulating fluidized bed reactor was developed around the sparger.
- The CO concentration within the fluidized bed passes through a maximum, and after converting the fuel to CO, the CO oxidation takes place along the reactor height as soon as it comes into intimate contact with oxygen.
- The results of this study show that the emission level at the reactor exit (i.e., CO and NO$_x$) is rather small and this leads to basic knowledge needed to develop novel heat-CO$_2$ integrated devices for heat generation and food-grade CO$_2$ production facilities in food and beverage industries. The purification cost and investment for such unit as compared with the catalytic reaction systems and

conventional devices is very competitive and would be considered as a framework to put forward the novel and friendly devices to protect the multiple issues and concerns related to the environment protection and global warming in food engineering and processing applications.

- The amount of sulfur in natural gas is usually very low. Even though, if in any case, the sulfur comes from the feed to the TFB reactor, one can add absorbents (i.e., limestone) to capture the SO_2 and very high capture efficiency can be insured with high gas-solid contact efficiencies expected in this reactor.

5. ACKNOWLEDGEMENTS

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6. NOMENCLATURE

a = alumina particles
D = reactor diameter (m)
d_p = mean particle size (μm)
F_{CH40} = initial methane molar flux (mol/s)
T = temperature (°C)
U = superficial gas velocity (m/s)
U_C = onset of turbulent fluidization (m/s)
U_j = jetting velocity (m/s)
X_{CH40} = initial methane mole fraction (-)
z = height along the bed (m)
τ = mean residence time (s)
η = contact time index (W/F_{CH40}) (g.s/mol)

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