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MODÉLISATION ET SIMULATION D'UN RÉACTEUR À LIT FLUIDISÉ CIRCULANT INTERNE POUR LE TRAITEMENT THERMIQUE DE DÉCHETS SOLIDES INDUSTRIELS

Par

Lukanda Samy MUKADI

DÉPARTEMENT DE GÉNIE CHIMIQUE

ÉCOLE POLYTECHNIQUE DE MONTRÉAL

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ÉCOLE POLYTECHNIQUE DE MONTRÉAL

Cette thèse intitulée :

MODÉLISATION ET SIMULATION D'UN RÉACTEUR
À LIT FLUIDISÉ CIRCULANT INTERNE
POUR LE TRAITEMENT THERMIQUE
DE DÉCHETS SOLIDES INDUSTRIELS

Présentée par: MUKADI Samy Lukanda
en vue de l'obtention du Diplôme de Philosophiae Doctor, Ph.D.,
a été dûment acceptée par le jury d'examen constitué de:

M. KLVANA Danilo, Ph.D. , ing., président
M. GUY Christophe, Ph.D., ing., membre et directeur de recherche
M. LEGROS Robert, Ph.D., ing., membre et codirecteur de recherche
M. ANTHONY Edward J., Ph.D., ing., examinateur externe
M. PERRIER Michel, Ph.D., ing., membre

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RÉSUMÉ

Le traitement de déchets industriels est devenu une nécessité de plus en plus pressante. Cela a entraîné le développement de plusieurs technologies qui sont actuellement disponibles. Cependant, face aux exigences environnementales et gouvernementales sans cesse plus sévères, le développement de procédés plus performants devient impératif. Les lits fluidisés, en général, possèdent plusieurs atouts pour le traitement thermique des déchets organiques solides finement divisés ou même des boues. Ils ont l'avantage de détruire la matière organique tout en produisant moins de polluants gazeux. Le Lit Fluidisé avec Circulation Interne, LFCI, renferme tous les atouts des lits fluidisés et a l'avantage d'être compact, englobant l'action de différentes unités opératoires: réacteur et séparateur gaz-solide. Il est utilisable comme unité mobile et est capable de traiter les déchets de différentes sources. Il offre, ainsi, une technologie prometteuse pour le traitement de déchets sur site, ce qui est un moyen pratique de réduire les coûts tout en satisfaisant aux exigences environnementales. Le présent travail se situe dans le cadre du développement d'un procédé LFCI pour le traitement thermique de déchets solides industriels.

Lors du traitement thermique, la destruction du déchet est généralement accompagnée de la production des gaz polluants nuisibles à l'environnement. Ainsi, la performance d'une unité de traitement thermique s'évalue par deux facteurs—le degré de destruction du déchet et la qualité des émissions gazeuses. Ces facteurs sont largement fonction des

variables telles que les conditions opératoires et caractéristiques géométriques. Une étude adéquate sur l'effet des ces variables permet le développement et l'opération optimale du LFCI. La modélisation et l'expérimentation sont des outils appropriés et complémentaires. La modélisation simule le fonctionnement du LFCI par un code informatique, lequel est utilisé pour étudier les effets de différentes variables sur la performance de l'unité. L'expérimentation sert d'une part à démontrer la faisabilité technique et d'autre part à valider la modélisation. Cette thèse a comme objectif la modélisation et la simulation du LFCI pour le traitement thermique de déchets industriels afin d'élaborer certains critères et stratégies utiles pour le design et l'opération d'une unité commerciale. Pour ce faire, la méthodologie suivante est considérée : (1) analyse du procédé afin de déterminer des hypothèses et des simplifications, (2) élaboration d'équations mathématiques basées sur les phénomènes d'échanges — hydrodynamique, transfert de chaleur et de masse ainsi que la cinétique de réaction, (3) résolution des équations, simulation et analyse des résultats, (4) validation à l'aide des observations expérimentales.

Un modèle mathématique prédictif est d'abord développé pour simuler principalement la conversion, la concentration de l'oxygène, le temps de séjour, la porosité et la vitesse des particules le long du tube de montée. Les paramètres d'entrée sont les débits du gaz et du solide, le taux de circulation ainsi que les propriétés physiques des différentes phases. Le modèle inclut un système de réaction très simple, constitué d'une seule réaction hétérogène. Les données expérimentales pour la validation de ce modèle proviennent du traitement thermique du sable de fonderie comme déchet représentatif. La validation de ce modèle se compare d'une manière satisfaisante aux observations expérimentales.

Ensuite, le modèle proposé est modifié pour calculer les concentrations des polluants gazeux, soit CO, CO_2, NOx, N_2O, SO_2 et O_2. Le système de réaction qui suppose la formation de volatiles et du char, comprend une vingtaine de réactions homogènes et hétérogènes. Ces réactions, tirées d'une recherche bibliographique approfondie, se rapportent au carbone, à l'azote, l'hydrogène, l'oxygène et le soufre contenus dans le déchet. Il est montré que les produits solides composant le lit fluidisé joue un rôle capital dans la formation et la réduction des émissions gazeuses. Le modèle hydrodynamique cœur-anneau est utilisé pour le tube de montée afin de tenir compte de l'effet des réactions rapides.

Enfin, le modèle qui a été validé pour le traitement de sable de fonderie, est utilisé pour faire une analyse paramétrique puis pour faire ressortir des critères ou stratégies nécessaires au design et à l'opération du LFCI. Les simulations sont faites pour analyser la performance du réacteur en présence de différents variables opératoires et géométriques ainsi que pour différentes propriétés physiques et chimiques du déchet. Les simulations démontrent que le taux de circulation, les débits du gaz et du solide, le diamètre du tube de montée, la température active dans le lit et les propriétés du déchet sont des variables qui affectent beaucoup la performance du LFCI. Ce travail fait ressortir un paramètre critique pour le design et l'opération— le rapport entre le débit du solide en circulation interne à celui du solide alimenté.

ABSTRACT

Treatment of industrial wastes has become an increasingly pressing need, which has led to the development of several treatment technologies in use today. However, the development of more performing processes becomes imperative due to the unceasingly stricter environmental and governmental regulations. In general, fluidized beds possess many benefits for thermal treatment of organic solid wastes, finely divided wastes or even sludge. They have the advantage of destroying the organic material while producing less gaseous pollutants. The Internally Circulating Fluidized Bed, ICFB, maintains all the characteristics of fluidized beds, and has the advantage of being shorter and more compact, combining some unit processes—reaction and gas-solid separation. It is favorable as a mobile unit, able to treat wastes of various sources. Therefore, it offers a very promising technology for on-site treatment as a means to reduce costs while meeting environmental requirements. This work lays within the framework of developing a process for the treatment of industrial solid wastes.

Destruction of waste through thermal treatment generally occurs with the generation of gaseous pollutants. Hence, the performance of any thermal treatment unit is best evaluated with two factors—the degree of waste destruction and the quality of gaseous emissions. These factors depend strongly on variables such as operating conditions and design parameters. Development and optimal operation of IFCB relays on proper investigation of the effect of different variables. In this respect, modeling and

experiments are appropriate and complementary tools. Modeling substitutes the ICFB operation with computer code, which then serves to study the effects of different variables. Experiment, however, is used to show technical feasibility and to validate the model. This thesis aims at modeling and simulation of the ICFB for thermal treatment of industrial waste in order to set out certain criteria and strategies useful for design and scale-up. Thus, the following methodology is employed: (1) analysis of the process in order to determine assumptions and simplifications, (2) development of mathematical equations based on the system's transport phenomena: hydrodynamics, heat and mass transfer as well as the kinetics of reaction, (3) solution of the equations including simulations and analysis of the results, (4) validation with experimental data.

A predictive mathematical model is initially developed to predict conversion, concentration of oxygen, residence time, voidage and sectional average particle velocity along the riser. Input parameters are feedrates of gas and solid, solid circulation flux and physical properties of different materials. The model is made with a very simple reaction system, which contains a single heterogeneous reaction. Experiment with spent foundry sand is undertaken in order to assess the validity of the model. The model predictions compare satisfactorily with experimental data.

Then, the proposed model is modified to calculate the concentrations of the gaseous pollutants, namely CO, CO_2, NOx, N_2O, SO_2, and O_2. The system of reaction, which assumes the formation of volatile and char, incorporates a number of homogeneous and heterogeneous reactions. An extensive literature review yielded reactions related to carbon, nitrogen, hydrogen, oxygen and sulfur. It is shown that bed materials play a major part in the formation and reduction of gaseous emissions. The core-annulus model is used for the riser to account for the effect of fast reactions.

Finally, the model which was validated for treatment of spent foundry sand, is used to make parametric analysis and set forth some approaches for an eventual scale-up. Simulations are performed to analyze the behavior of the reactor against different operating and design variables as well as waste properties. Simulations reveal that the variables most affecting the ICFB performance are solid circulation flux, gas and solid flowrates, riser diameter, working bed temperature and waste properties. In this work, the ratio of solid circulation flowrate to feedrate is recognized as a critical parameter for design and operation.

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Nomenclature du Chapitre 2

A : section, or contact area, m^2

C_D : drag coefficient

C_p : constant pressure heat capacity, J/kg/K

Deff: effective diffusivity m^2/s

d_p , d_r, D_w : particle, riser, column diameter, m

E : activation energy, J/mol

$F_{g\text{-}w}$, $F_{s\text{-}w}$: gas and solids frictional forces, Pa/m

F_w: shape factor

Fr, Fr_t : Froude number

g : gravity acceleration, 9.8 m/s^2

Gs : solids circulation flux, kg/m^2s

ΔH : heat of reaction, J/kg_{resin}, J/kg (CH_4)

h : heat transfer coefficient, $W/m^2/K$

H_a, H_c : annular and cylinder height, m

ko : pre-exponential factor

k_{O2}, k_r : reaction rate constants

K: constant

Lacc : length of acceleration zone, m

m_g, m_s : mass of gas and solids, kg

M_m: molecular mass kg/kmol

$n1$, $n2$: oxygen and resin reaction order

p_{O2} : oxygen partial pressure, Pa

P_{tot}: reaction pressure, 1.013×10^5Pa

Q_{loss} : heat losses through walls, W

r : rate of reaction, $kg_{resin}/kg_{sand}/s$

ri: inner refractory radius, m

ro: outer cylinder radius, m

R : universal gas constant, 8314.4 J/kmol.K

Re_p : particle Reynolds number

T : temperature, K

t : time, s

Ut : terminal velocity of sand, m/s

Uo : superficial gas velocity, m/s

V_p : particle sectional average velocity, m/s

W : flowrate, kg/s

x : conversion

Y_p : resin weight fraction

Y_{O2} : oxygen molar fraction

z : axial position, m

Greek symbols

α : absorbtivity

ρ : density, kg/m^3

ξ: emissivity

ϕ : slip factor

σ : Stephan-Boltztman constant, $5.67\times10^{-8}W/m^2/K^4$

μ : viscosity, Pa.s

ε : voidage

τ: residence time,s

υ_{O2} : stoichiometric coefficient of oxygen

Subscripts

∞ : fully developed

air : air

a : annular bed

f : disengagement zone

feed : feed

g : gas

nat : natural gas

o : outer

p : particle

r : riser

tot : total

w : wall

Nomenclature du Chapitre 3

$C_{a,i}$: "i" species annulus concentration, kmol/m^3

$C_{c,i}$: "i" species core concentration, kmol/m^3

D: riser diameter, m

D_c: core diameter, m

D_v: diffusivity, m^2/s

F_i: molar flowrate, kmol/s

Fr: Froude number

g: gravity acceleration, 9.8 m/s^2

Gs: solids circulation flux, kg/m^{2}s

Kg: gas interchange coefficient, m/s

k_i: "i" species rate constant

P: pressure, Pa

r : radial position, m

r_c: core radius, m

R: riser, m

R_g: gas constant, 8314.4 J/kmol.K

R_i: "i" species reaction rate: kmol/m^3

T: temperature, K

t: time, s

U_g: superficial gas velocity, m/s

v: volumetric flowrate, m^3/s

V_D: disengagement zone volume, m^3

Y_i: molar fraction

z: position, m

ρ, ρ_p: gas and particle density, kg/m^3

φ: sectional ratio

μ: viscosity, Pa.s

ε: voidage

Nomenclature du Chapitre 4

A_r: riser section, m^2

G_s: solids circulation flux, kg/m^2s

$m_{s,tot}$: total mass of solids, kg

n: aeration rate

N_{pass}: number of recycles

$R_{s,i}$: reaction/drying rates, kg/m^2s

t: time, s

V_p: sectional average particle velocity, m/s

W_{air}: air mass flowrate, kg/s

W_{feed}: waste feedrate, kg/s

Y_{pr}: organic mass fraction

z: position, m

Greek symbols

ε: voidage

ρ_p: solids density, kg/m^3

Chapitre 1___INTRODUCTION GÉNÉRALE

1.1 Introduction

Le monde industriel génère quotidiennement de grandes quantités de déchets qui nécessitent un mode de disposition acceptable pour l'environnement. Bien que les modes traditionnels d'enfouissement et d'entreposage restent encore répandus, leur simple usage n'est plus totalement approuvé. Les normes et les réglementations gouvernementales de plus en plus sévères, exigent un traitement avant enfouissement ou entreposage de certains types de déchets. De plus, le recyclage et la valorisation de la matière contenue dans les déchets se révèlent comme des alternatives économiquement rentables et permettent d'éviter la pollution des nouveaux sites. Ainsi, le traitement des déchets devient une nécessité de plus en plus pressante sinon une obligation.

Ceci a entraîné le développement de plusieurs technologies de traitement de déchets industriels telles que traitements chimiques, physiques, thermiques et biologiques [Freeman, 1998]. Par contre, la sélection d'une technologie quelconque dépend beaucoup de la nature physico-chimique du déchet. Ainsi, pour le type de déchets industriels d'origine organique, finement divisés ou des boues, le traitement thermique par combustion en lit fluidisé offre des nombreux avantages par rapport aux technologies traditionnelles basées sur les fours rotatifs, les foyer fixes, les foyers multiples, les chambres multiples, ou même le procédé gaz-contact [Freeman,1998, Dempsey et Oppelt, 1993; Kaferstein et al., 1997, Benali et al., 1992a,b,c].

Des procédés à fours rotatifs par exemple, ont l'avantage de traiter une large gamme de déchets solides. Cependant, ces procédés ont tendance d'être de grande taille, utilisent trop d'excès d'air, ont un mauvais contact entre les gaz et la matière solide et les fumées de combustion générées contiennent souvent d'importante concentration de polluants (CO, NOx, N_2O, SO_2, imbrûlés), alourdissant ainsi le fardeau du procédé de post-traitement des fumées. Le procédé gaz contact [Benali et al., 1992a,b,c], bien qu'efficace pour le traitement de certains déchets solides, est défavorisé par la difficulté de contrôler le temps de séjour du solide qui dépend seulement de la longueur du lit, vu que le solide tombe par gravité.

Le lit fluidisé de son coté, a l'avantage – de pourvoir un bon contrôle du temps de séjour – d'opérer à une température uniforme propice pour le contrôle, –d'avoir une bonne inertie thermique, – et de favoriser le contact intime entre les différentes phases ce qui est responsable de taux rapides de transfert de chaleur et de masse. Ils trouvent plusieurs applications industrielles dans les opérations impliquant le procédé gaz-solide telles que les procédés catalytiques, la combustion du charbon, de la biomasse et des déchets. En outre, une revue de la littérature démontre que la matière solide du lit fluidisé joue un rôle prépondérant dans la formation et la destruction des principaux polluants gazeux comme les oxydes d'azote et de soufre [Anthony et Preto 1995; Bulewicz et al., 1997; Desai et al., 1995; Johnsson et Dam-Johansen, 1995; Johnsson et al., 1997; Goel et al., 1994; Orlanders et Stömberg, 1995; Winter et al., 1996 et 1997; Li et al., 1998]. En général, le traitement en lit fluidisé permet facilement la destruction de déchets solides et une faible émission de polluants gazeux.

Le Lit Fluidisé à Circulation Interne (LFCI) a été développé pour résoudre le problème du temps de séjour du gaz dans le lit à jet [Milne et al., 1992]. Étant composé d'un tube de montée et d'un tube externe contenant le lit, le LFCI diffère du lit à bulles et possède toutes les caractéristiques d'un lit fluidisé circulant [Figure 1.1]. La présence du tube de montée sert à limiter l'écoulement du gaz à l'intérieur du tube en plaçant le distributeur à gaz directement en bas du tube. Le tube de montée opère en mode de fluidisation rapide et possède une grande porosité. Ce système permet d'isoler et d'opérer une partie du réacteur à une température beaucoup plus grande que le reste du lit; cela est assuré par l'utilisation d'un brûleur et le maintien facile d'une flamme distincte dans le tube de montée. Ceci est très difficile à réaliser dans un lit à bulles à cause de la densité du solide dans le lit. Les taux de transferts élevés permettent d'avoir une unité plus compacte, appropriée comme unité mobile, qui combine les étapes de réaction et de séparation directe gaz-solide par la présence d'une zone de désengagement au-dessus du tube de montée et l'inclusion d'une plaque d'impact. Ceci est une technologie prometteuse pour le traitement de déchets sur site, qui est un moyen pratique de réduire les coûts tout en satisfaisant aux normes environnementales [Baukal et al., 1994; McGowan et Ross, 1991].

Un design efficace et une stratégie d'opération adéquate sont requis pour une telle unité qui peut être assujettie à des conditions transitoires variables et à différents types de déchets. Il est donc impératif de développer un modèle capable de décrire et de caractériser la performance du LFCI pour le traitement thermique sous différentes conditions opératoires et paramètres géométriques. Le travail expérimental à permis d'étudier la faisabilité technique du procédé pour le traitement des déchets industriels [Mukadi et al., 1997, Boisselle et al., 1998]. La modélisation combinée au travail expérimental permet de réaliser une étude beaucoup plus approfondie. Elle permet de simuler et d'analyser la performance du réacteur face à la variation de plusieurs conditions opératoires et géométriques. Ceci sert ensuite à élaborer des critères et des stratégies utiles pour le design ou l'extrapolation à l'échelle industrielle.

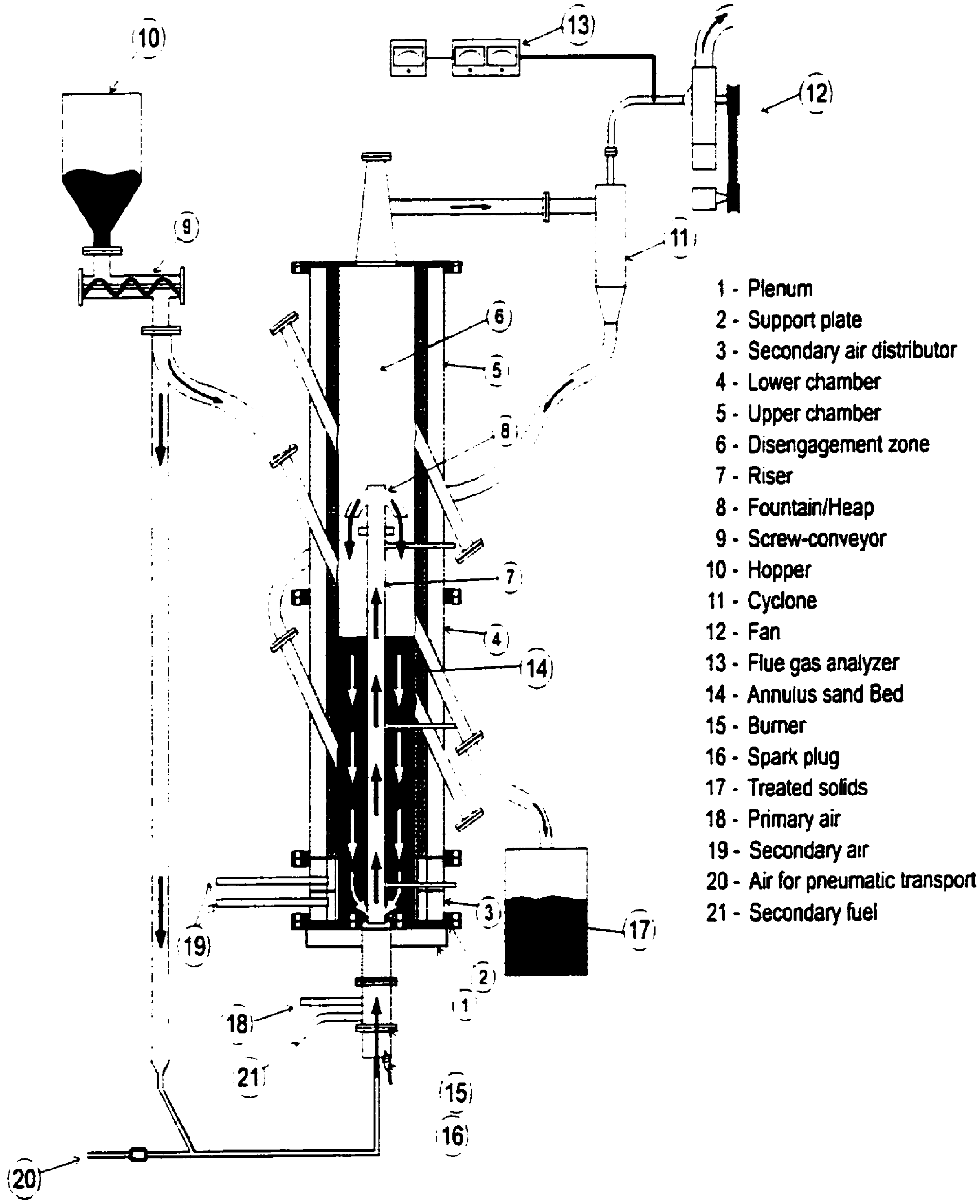

Figure 1.1 : Unité expérimentale pour le traitement thermique de déchets à lit fluidisé circulant interne.

1.2 Objectif

Le présent travail se fixe les objectifs suivants:

- Modélisation mathématique du LFCI basée sur l'hydrodynamique, le transfert de chaleur et de masse ainsi que les cinétiques des réactions présentes.
- Simulation de la conversion du déchet, des concentrations des émissions gazeuses et des profiles de température.
- Élaboration de certains critères et stratégies pour le design et l'opération.

1.3 Méthodologie

Lors du traitement thermique, la destruction du déchet est toujours accompagnée de la production des gaz polluants. Ainsi, la performance du LFCI est évaluée par le degré de destruction de la matière organique et la qualité des émissions gazeuses. La méthodologie est composée d'une approche théorique et expérimentale. L'approche théorique sert à analyser les phénomènes physiques et chimiques du système, à développer des hypothèses et simplifications relatives à la phénoménologie du réacteur. Ceci aboutit à l'élaboration des équations mathématiques caractérisant le réacteur. L'expérimentation est utilisée pour tester certaines hypothèses et valider le modèle en comparant les valeurs prédites aux valeurs expérimentales. Cette méthodologie est résumée à la figure 1.2.

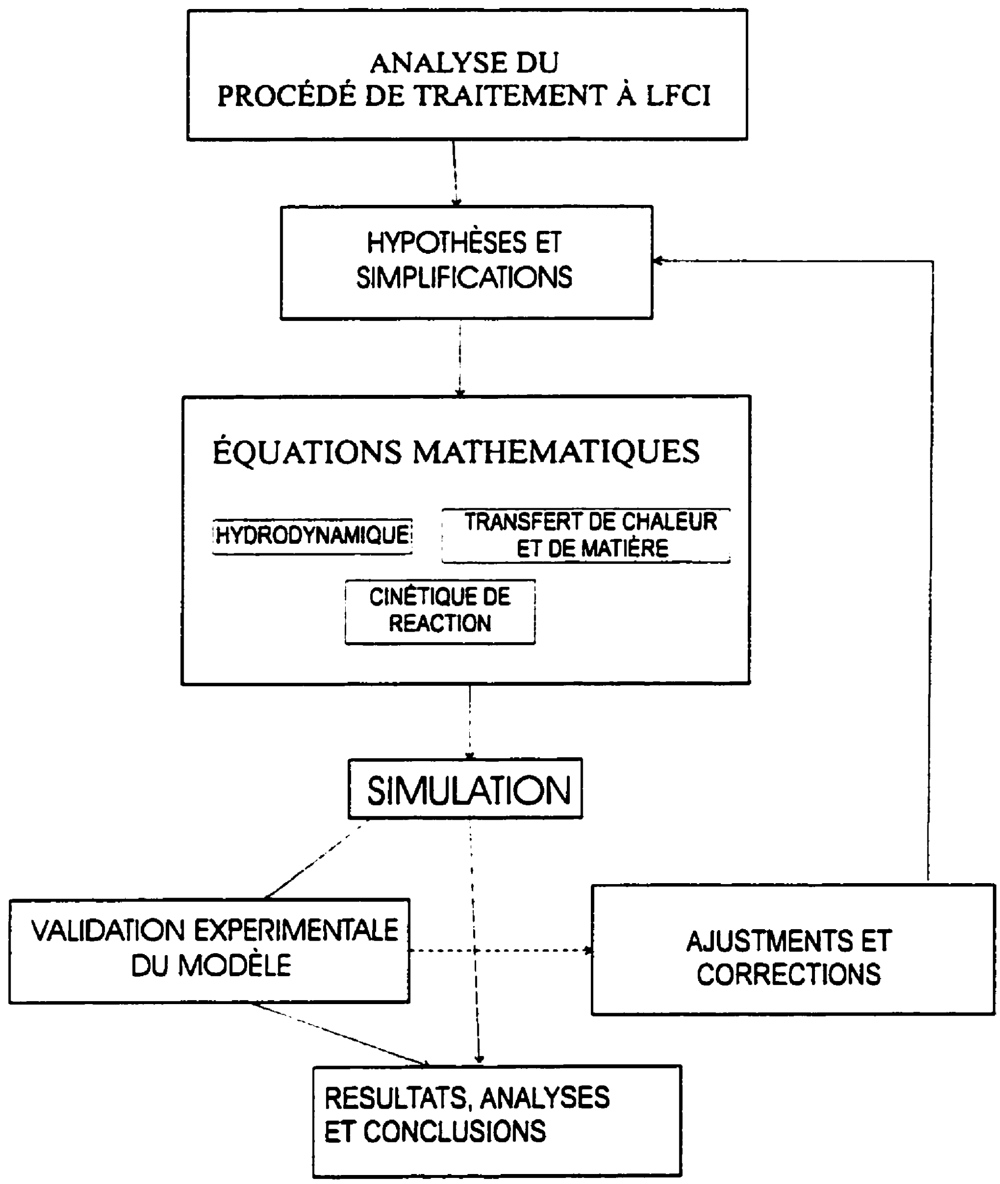

Figure 1.2: Schéma descriptif de la méthodologie utilisée durant la modélisation

1.4 Structure de la thèse

Cette thèse comprend trois articles :

Chaque article inclus une revue de la littérature pertinente en abordant le problème selon la méthodologie suggérée précédemment. La thématique de fond de chacun des articles porte sur le développement et le design du LFCI pour le traitement thermique des déchets industriels tels que les sables de fonderie, les sols contaminés, les électrodes de l'industrie d'aluminium, les boues procédés (industrie des pâtes et papier, traitement des eaux usées, fonds de réservoirs de pétrole, etc.) et d'autres déchets solides finement divisés. Cependant le sable de fonderie est sélectionné comme déchet représentatif pour l'étude de la modélisation.

Le premier article, chapitre 2, traite la modélisation et la simulation de déchets solides en considérant tous les phénomènes d'échanges dans le réacteur. Le système réactionnel suppose que les réactions en phase gazeuse sont très rapides et instantanées par rapport à la combustion du solide; ainsi une réaction globale est considérée pour prédire les températures, la conversion de la matière organique du déchet solide et la concentration de l'oxygène.

Le second article, chapitre 3, se rapporte aux émissions gazeuses. Il contient un système de réactions homogènes et hétérogènes dérivées dans la littérature traitant la formation et la réduction des polluants gazeux dans les lits fluidisés. Ces réactions sont normalement très rapides et n'ont pas d'effets significatifs sur l'approche utilisée au chapitre deux.

Le troisième article, qui se trouve au chapitre 3, fait l'objet d'une étude paramétrique en utilisant le modèle proposé dans le chapitre 2. Il montre la capacité de prédiction suite à la variation du type de déchet, des conditions opératoires et géométriques en indiquant la présence des paramètres critiques pour le design et l'opération.

La partie « conclusions et recommandations » reprend les principales contributions de ce travail ainsi que les suggestions pour de futures recherches. Dans les annexes se trouvent différents schémas, tableaux et graphiques complémentaires.

Chapitre 2___MODELING OF AN INTERNALLY CIRCULATING FLUIDIZED BED REACTOR FOR THERMAL TREATMENT OF INDUSTRIAL SOLID WASTES

Reference: Lukanda Mukadi, Christophe Guy* and Robert Legros (1998): "Modeling of an Internally Circulating Fluidized Bed Reactor for Thermal Treatment of Industrial Solid Wastes", Submitted to *Canadian Journal of Chemical Engineering*.

Keywords: Circulating Fluidized Bed, Combustion, Solid Waste, Modeling

* Author for correspondence, E-mail address: Christophe.Guy@mail.polymtl.ca

2.1 Abstracts

2.1.1 Abstract

A predictive mathematical model is developed to describe and characterize the key design variables of a novel Internal Circulating Fluidized Bed Combustor. The model is based on fundamental principles of heat transfer, mass transfer, hydrodynamics and reaction kinetics. Under justifiable assumptions unsteady state mathematical equations are written and solved. The input parameters of the model are gas and solid flowrates, solid circulation flux, as well as physical properties of gas and solids. The model allows to predict and to investigate the temperature, the combustible conversion, the oxygen concentration, the residence time, the voidage and the solid velocity in the riser. Experiments with spent foundry sand are undertaken in order to assess the validity of the model. The predictions for the transition period of heating clean sand and the treatment of waste compare satisfactorily to the experimental data.

2.1.2 Résumé

Un modèle mathématique prédictif est développé pour décrire et caractériser les paramètres importants dans la conception d'un nouveau Réacteur à Lit Fluidisé Circulant Interne. Le modèle est basé sur les principes fondamentaux de transfert de chaleur, transfert de masse, l'hydrodynamique et la cinétique de réaction. En posant des hypothèses justifiables, des équations mathématiques du régime transitoire sont écrites et

résolues. Les paramètres d'entrées au modèle sont : les débits du gaz et du solide, le taux de circulation ainsi que les propriétés physiques des différentes phases. Le modèle permet de prédire la conversion, la concentration de l'oxygène, le temps de séjour, la porosité et la vitesse des particules solides dans le tube de montée. Les données expérimentales pour la validation du modèle proviennent du traitement thermique du sable fonderie comme déchet représentatif. Les prédictions du modèle et les résultats expérimentaux se comparent d'une manière satisfaisante pour la période de préchauffage du sable propre et le traitement du déchet.

2.2 Introduction

2.2.1 Introduction

Thermal treatment of wastes by combustion is becoming an important means for recycling, energy recovery or destruction of industrial solid wastes and sludge. The design of more efficient reactors is necessary to improve the performance of current treatment technologies and to develop new processes. For particulate wastes, a fluidized bed reactor presents the best combustion environment (Wilbourn et al., 1986; Mullen, 1992; Saxena and Rao, 1993; Sadhukhan and Bradford, 1993) as it offers a prime combination of temperature, residence time and turbulence. Bubbling and circulating fluidized beds are extensively used in biomass and coal technology (Kim et al., 1997; Kaferstein et al., 1997; Koskinen et al., 1995; Legros et al., 1995; Davidson, 1992). Much effort is still devoted to the study of thermal treatment of waste under fluid bed conditions (Li et al., 1997; Arena and Cammarota,1997; Dervin et al., 1997; Desai et al., 1995; Kozinski et al., 1995). As is the case for coal combustion, circulating fluidized beds offer enhanced performance in terms of waste destruction, low emissions and energy recovery. A novel Internally Circulating Fluidized Bed reactor (ICFB) is based on a flame-solid direct contact process to treat industrial waste (Guy et al., 1997; Mukadi et al., 1997). A burner located at the riser base provides a region of intense flame-solid contact and allows high treatment temperature in the presence of radicals; this is advantageous for rapid devolatilization and combustion. Benali et al. (1992 a,b,c) developed a flame-solid direct contact process with a down-flow reactor in which residence time and conversion are difficult to control because solids flow by gravity. Wastes that require longer residence time, such as humid wastes or large granular solids cannot be adequately treated. The novel ICFB Reactor circumvent these pitfalls. The solids residence time is simply controlled via the solids flowrate and the total inventory of solids in the reactor. Moreover, the presence of a draft-tube-riser procures a region of lower solid density. This low density region is suitable to maintain and confine a distinct flame which is difficult to achieve in most bubbling and turbulent beds due to their high bed density. The essence of the flame-solid contact process is thus maintained. A detailed description of the process has been presented in the patent application (Guy et al.,1997) and technical and economical feasibility has been proven for foundry sand reclamation (Mukadi et al.,1997). This paper presents the modeling and simulation of the ICFB reactor for design purposes.

2.2.2 Industrial Wastes

Many industrial wastes generated today require treatment to meet environmental regulations. The present work relates to waste containing organic material and having low heating value, such as : spent foundry sand, contaminated soil, electrodes from the aluminum industry, pulp and paper de-inking sludge, sludge from waste water purification, sludge from petroleum reservoirs, industrial sludge, and other divided solid wastes. Spent foundry sand has been selected as a representative industrial waste in this study on ICFB reactor modeling. It has the advantage of being a well characterized and uniform waste (Reier, 1993; Lessiter, 1994; Philbin, 1995; Bralower, 1989) . Foundry sand comes from the mixture of clean sand with a binder, used to make mold for metal casting. Foundry sand may be reused several times before it loses its molding capacity and becomes spent foundry sand. In the latter form, it is coated with a degraded binder which must be removed to reclaim the sand grains. Typical spent foundry sand contains less than 5% of binder, whose chemical composition depends on both the foundry and the casting process.

2.2.3 Experimental procédure

The ICFB apparatus and dimensions are presented in Figure 2.1. The reactor is composed of two concentric cylinders and a natural gas burner vertically located at the base of the inner tube called the "riser". Located on top of the riser is the gas-solid separation device and the disengagement. Particles from the disengagement zone fall into the annular space where they flow downwards. Secondary air is used at the base of the outer tube to prompt solid motion and to set solid circulation rate. Solids from the annular space re-enter the riser through perforated holes at the riser bottom. The solid feed-system is composed of a screw-conveyor and pneumatic transport section. Solids can be fed through the burner at the bottom of the riser, in which case pneumatic air is used to transport solids into the reactor, or they can be fed directly into the annulus region. Fluidizing gas enters the bed through the burner at the riser base. Particles from the annulus and fresh waste are entrained and carried up into the fountain zone. Orifice flowmeters are used for air and natural gas. Measurements of temperature and pressure as well as solid sample withdrawals are performed at different axial positions of the reactor (3 positions for the riser and 5 for the outer cylinder). Type B thermocouples with ceramic shields are used to measure temperatures in the riser. Unshielded type K thermocouples are used for all other temperature measurements. Concentrations of NO_x, CO, CO_2, SO_2, O_2 and unburned hydrocarbons are measured with an on-line gas analyzer. Pressure drops through the orifice flowmeters and pressure profiles within the reactor are measured with pressure transducers. Data acquisition is provided by an on-line computer.

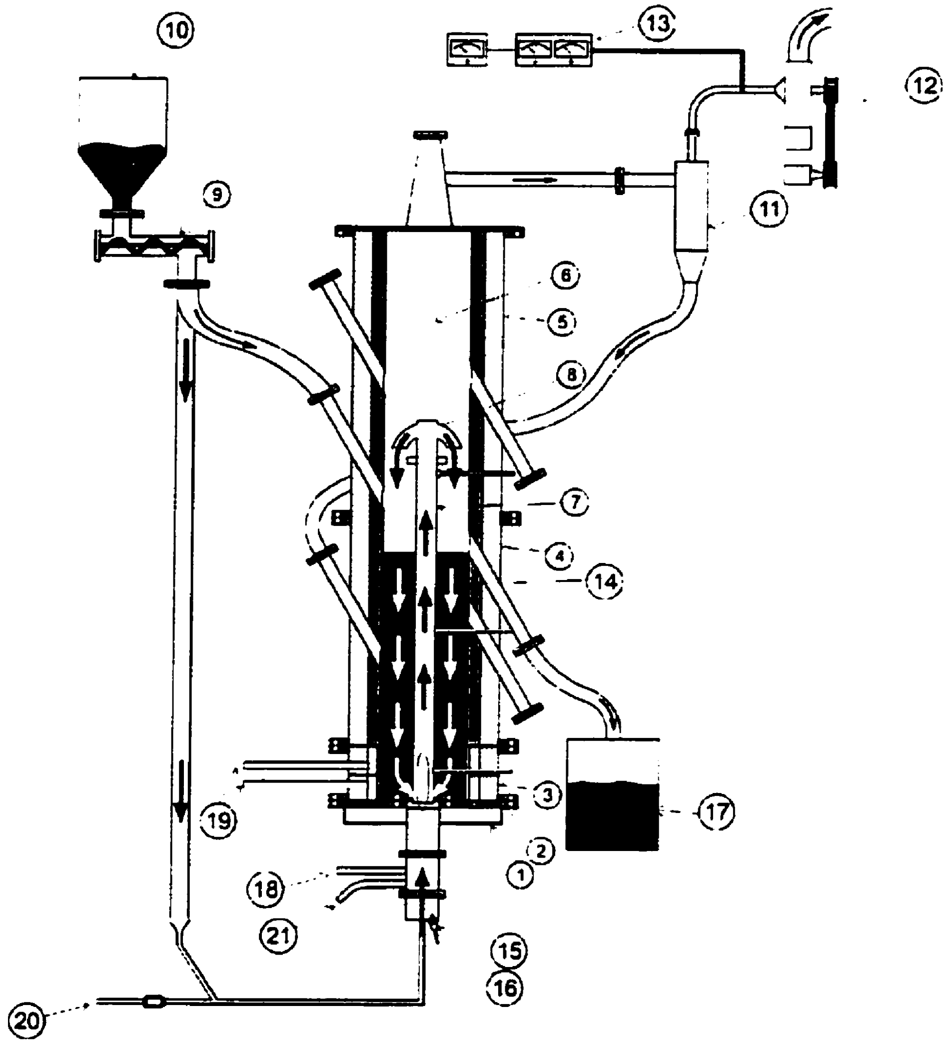

Figure 2.1: Schematics of the Internally Circulating Fluidized Bed pilot unit: 1-Plenum, 2-Support plate, 3- Secondary air distributor, 4-Lower chamber, 5-Upper chamber, 6-Disengagement zone, 7-Riser, 8-Impact device, 9-Screw-conveyor, 10-Hopper, 11-Cyclone, 12-Ventilator, 13-Flue gas analyzer, 14-Annulus region, 15-Burner, 16-Spark plug, 17-Treated solids, 18-Primary air, 19-Secondary air, 20-Air for pneumatic transport, 21-Natural gas. . Riser --length: 1.5m/diameter: i.d. 780 mm, o.d. 890 mm. External cylinder-- length: 2.5m/ diameter: i.d. 355 mm, o.d.645 mm. Burner length: 300 mm..

2.3 Circulating Fluidized Bed Reactor Models

The ICFB is similar in many ways to the traditional CFB. The main difference is in the mode of solid circulation. In the ICFB, solids circulate through the annular zone whereas in the CFB solids circulate through a standpipe. The riser, the annular solids bed and the fountain zone of the ICFB are basically equivalent in function as the riser, the standpipe and the cyclone of the traditional CFB. The ICFB is more compact and heat losses are likely to be lower since its riser is not exposed and its annular bed acts as an additional refractory. Milne et al. (1992) have also presented the ICFB as a modification of a spouted bed.

Extensive work has been done to model the traditional CFB no ICFB models exist. Accurate reactor modeling requires hydrodynamics, heat and mass transfer as well as reaction kinetics in order to predict reactor performance. Several CFB modeling approaches lead to models with different degrees of sophistication (Grace and Lim, 1997; Hyppanen et al., 1991; Gidaspow, 1994). While the cyclone, downer or annulus are commonly treated as well mixed or plug flow systems; the riser modeling varies a lot — from homogeneous to heterogeneous modeling, from single-region to multiple-region modeling and from 0D to 3D modeling or a combination of these approaches.

Single-region pseudo-homogeneous models assume that hydrodynamic radial (and axial) gradients are negligible. Hastaoglu et al. (1988), Gianetto et al. (1990), Ouyang et al. (1993) adopted models for which both gas and solids are in plug flow through the entire riser while ignoring axial variations in voidage and other hydrodynamic properties. These assumptions are acceptable for taller risers where the height of the bottom zone is negligible. Also, Lee and Hyppanen (1989), Pagliolico et al. (1992) and Marmo et al. (1995) presented models in which the riser acts as a PFR for the gas and a CSTR for the solids, and the axial voidage profile in the riser is considered. These type of models are a good approximation for lower density pneumatic transport reactors and also for risers with smaller L/D ratios (Contractor et al. ,1994; Marmo et al., 1995; Weiss and Fett, 1986; Muir et al., 1997).

Some models approximate axial variation of variables by dividing the riser into several cells in series, each with different voidage, and different mixing characteristics of solids and gas. Legros et al. (1995) and Marmo et al. (1995) divided the riser into 2 parts with the lower dense region being considered as a perfectly mixed turbulent bed rather than in plug flow. The solids are considered either well-mixed (Arena et al., 1995), in plug flow (Muir et al, 1997; Saraiva et al., 1993) or in bubbling bed mode (Saraiva et al., 1993; Zhang et al., 1991).

Core-annulus models have been developed to account for radial non-uniformity in the riser (Bolton and Davidson, 1988; Senior and Brereton, 1992; Kagawa et al., 1991; Patience and Chaouki, 1993; Puchyr et al., 1997). These models assume that all of the gas and some solids pass upwards through a central core, and that the gas and solids are being exchanged with an outer annular region where the gas and the solids are stagnant or flow downwards. Marmo et al. (1995) modeled the ozone reaction in the riser; the lower dense zone was considered to be in the bubbling regime.

2D and 3D models have also been developed for CFB (Schoenfelder et al., 1996; Balzer and Simonin, 1997). Despite the improvement of CFB models through increasing degree of sophistication, the lower dense region is still difficult to model. This lower dense zone has been treated as a turbulent bed (Talukdar et al., 1994), a bubbling bed (Marmo et al., 1995; Neidel et al., 1995; Saraiva et al., 1993), an acceleration zone (Pugsley et al., 1992) or a combination of bubbling-acceleration zone (Neidel et al., 1995). Stoker et al. (1989) modeled a draft tube spouted bed with an approach valid for higher superficial velocities and higher voidage. The existing models can not be directly applied to shorter CFB reactors where the lower dense zone is the principal part of the reactor. This paper presents such a model for a short Internally Circulating Fluid Bed Combustor.

2.4 Reactor Model Development

The reactor under study is operated by first heating a fixed amount of clean sand, kept in batch, until steady state temperatures are reached. At this time, solid waste is continuously fed through the burner at the riser base and withdrawn from the annulus by overflow. For a constant feed, temperatures and concentration will change to reach a new steady state. A conservative reactor model must account for both radial and axial variations of design variables. The need for this model is important when flow non-uniformity exists or when the intrinsic reaction rate is rapid. Such is the case of several gas phase reactions in CFBs. In the present ICFB reactor model, flow non-uniformities are characterized by dividing the bed into hydrodynamically distinct zones : the riser, the annulus and the fountain zone as shown in Figures 2.2. Conservation equations of each distinct zone are written for the gas and the solid phases. As it is shown in the following sections, these equations are based on the hydrodynamics, heat and mass transfer as well as the reaction kinetics.

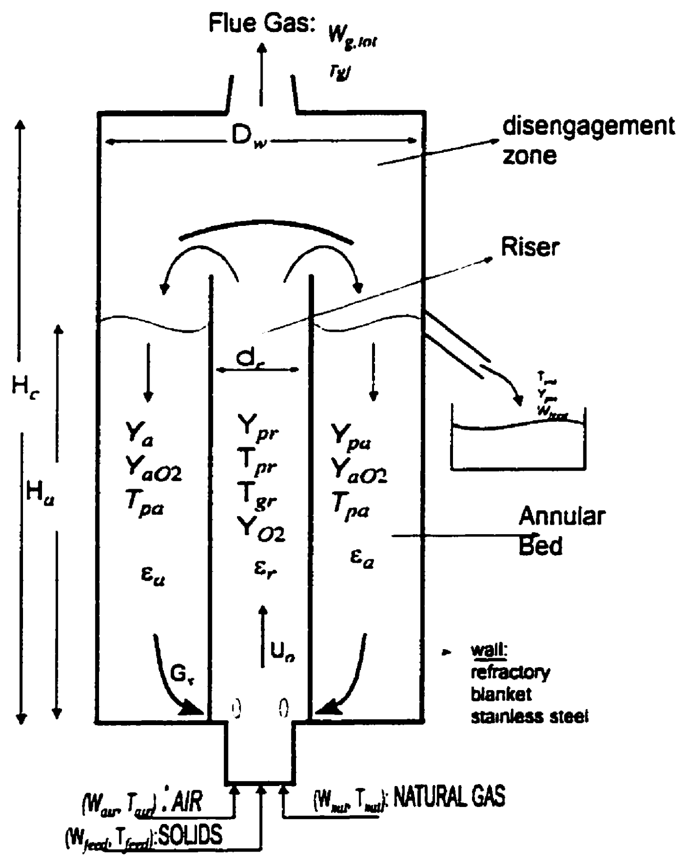

Figure 2.2: Schematic flow diagram of the ICFB Reactor

2.4.1 Kinetic Model

When compared to other wastes, foundry sand is well characterized but its resin is still a complex material whose chemical composition and reaction mechanism are not well known. This resin has lost most of its volatile during the high temperature metal casting process. Table 2.1 presents important properties of spent foundry sand. Non-isothermal thermogravimetric analysis is effective for studying the pyrolysis and combustion mechanisms of such complex substance as biomass and coal (El-Kalyoubi and El-Shinnawy, 1985; Tia et al., 1991a,b; Encinar et al., 1995; Chang et al., 1996). Thermogravimetric analysis (TGA) provides a measurement of the weight loss of the sample as a function of time and temperature. Under well controlled conditions, the problems related to transport phenomena are minimized by use of fine particles and small sample size (Haines, 1995). TGA was used for spent foundry sand resin to determine a simple kinetic model for engineering purposes. The combustion of foundry sand resin was studied over temperatures of 100 to 1000°C at different oxygen concentrations. Pure nitrogen was mixed with air to obtain different oxygen concentrations. A data-processing computer recorded data at a sampling rate of 1 Hz. This procedure is described elsewhere (Haines, 1995). Typical experimental data are presented in Figure 2.3. Following Wendlandt (1974) and Encinar et al. (1995) an Arrhenius formulation is used to express the kinetic model as:

$$r_{waste} = Y_{p,feed}\frac{dx}{dt} = k_0 e^{-E/RT} p_{O2}^{n1}(1-x)^{n2} \tag{1}$$

Where $Y_{p,feed}$ is the initial resin concentration in the sample, x is the resin conversion at time t, k_0 is the pre-exponential factor, E is the activation energy, T is the absolute temperature, R is the universal gas constant. Data such as in Figure 2.3 were directly fit to Equation 1 by non-linear regression method of Marquardt (1963, in Dennis and Schnabel, 1983). The activation energy was between 100 to 134 kJ/mol; the order, n2, varied from 1.85 to 2.2. Average values of the activation energy and the order 'n2' are taken as 120.6 kJ/mol and 2 respectively. Subsequently the oxygen order was found to be 1.3. Similar values have also been reported for the combustion of other solids fuels, biomass and coal (Encinar et al, 1995; Chang et al, 1996). The kinetics model is expressed as:

$$r_{waste} = 2.733 \times 10^5 \, e^{-14500/T} \, Y_{O_2,s}^{1.3} \left(\frac{Y_p}{Y_{p,feed}} \right)^2 \quad [\equiv] \frac{kg_{resin}}{kg_{sand} \cdot s} \tag{2}$$

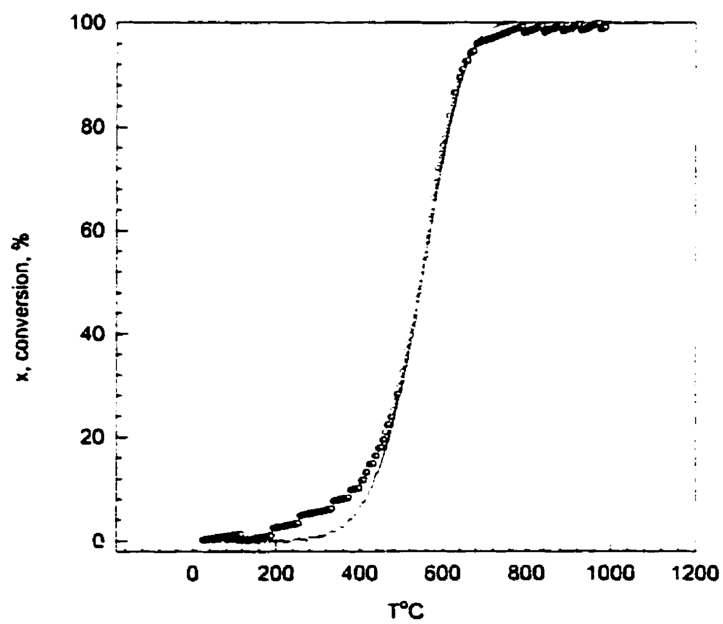

Figure 2.3: Combustion Kinetics of Foundry Sand Resin: Comparison between Computed and Experimental Data at atmospheric pressure and oxygen partial pressure, P_{O2}=10.64 kPa

The model fits well the experimental data as shown in Figure 2.3. The under-estimation of the model at lower conversion and lower temperature is not important because most of the combustion process occurs at higher temperatures where the model is more accurate. The corresponding reaction rate of oxygen is found by assuming an empirical chemical formula for the waste as: $C_c H_h N_n S_s O_x$, where the atomic quantities (c, h, n, s, x) of each element are obtained from the ultimate analysis in Table 2.1. The stoichiometric reaction is written as:

$$C_c H_h N_n S_s O_x + \upsilon_{O2} O_2 \longrightarrow c\, CO_2 + n\, NO + \frac{h}{2} H_2O + s\ SO_2 \tag{3}$$

with: c=149.63 , h=122.01, n=6.0675, s=1, x=34.747 and molar weight, M_{mwaste}=2590.5kg/kmol. The stoichiometric coefficient, υ_{O2}, and the reaction rate of oxygen are given by:

$$\upsilon_{O2} = \frac{1}{2}(2c + n + h/2 + 2s - x) = 166.8 \tag{4}$$

$$r_{O2} = \frac{\upsilon_{O2}}{M_{m,waste}} r_{waste} = 0.0644\, r_{waste} \quad [\equiv] \frac{kmole, O_2}{kg\ sand.s} \tag{5}$$

Table 2.1: Foundry sand properties, Proximate and ultimate analysis of resin

Physical Properties	Proximate analysis, wt%	Ultimate analysis, wt%
Sand type: Silica sand	Resin concentration:2.06	Carbon: 1.56
Particle mean diameter: 250 μm	Moisture :0.19	Hydrogen: 0.106
Combustion heat: 26MJ/kg resin	Volatile :<<0.01	Oxygen: 0.483
	Ash : 97.75	Nitrogen :0.0738
		Sulfur: 0.0278

2.4.2 Hydrodynamic Model And Analysis

The hydrodynamic modeling is treated according to the distinct zones shown in Figure 2.2. The solid circulation flux, G_s, and the superficial gas velocity are important design parameters for both modeling and operation. They influence particle residence time, solid holdup in the riser and temperature profiles in the reactor. At high values of G_s, solid phase behavior in the reactor would resemble complete mixing (CSTR). At low superficial gas velocity and higher circulation flux, choking or flame extinction can occur. The flowing gas transport the particles which enter the riser with velocity of zero. Because the riser is short, transported particles are constantly under acceleration and riser voidage varies with axial position.

2.4.2.1 Riser

The riser is assumed to be in pseudo-plug flow; the modeling approach follows the core-annulus model proposed by Pugsley and Berruti (1996). The zone between the riser and the impact device is considered as an extension of the riser. The momentum balance on particles is written as:

$$V_p \frac{dV_p}{dz} = \frac{3}{4} C_D \frac{\rho_g}{d_p \rho_p} \left(\frac{U_o}{\varepsilon} - V_p \right)^2 + g \frac{\rho_g - \rho_p}{\rho_p} \; ; \tag{6}$$

$$\text{with} \qquad V_p|_{z=0} = 0 \; ; \qquad \text{and } W_s = G_s A_r = \rho_p V_p A_r (1 - \varepsilon) \tag{7}$$

V_p is the cross sectional average upward particle velocity. The parameter C_D is determined by a correlation similar to that used for the terminal velocity of a single sphere in a fluid:

$$C_D = \frac{K}{Re_p^{0.6}}; \qquad Re_p = \frac{\rho_g d_p \left(U_o/\varepsilon - V_p\right)}{\mu} \tag{8}$$

In the acceleration regime, Re_p remains in the Stokes regime (Re_p<100), and the parameter K is obtained by using the boundary condition at the end of the acceleration zone when Vp becomes a constant (dVp/dz=0) equal to the velocity in the fully developed zone:

$$\text{at } z = L_{acc}\ ;\ V_p = V_{p\infty}\ ;\ \frac{dV_p}{dz} = 0 \tag{9}$$

Applying this limiting condition allows the calculation of K as a function of Uo and Gs. In the fully developed zone, the average solids upflow velocity ($V_{p\infty}$) and the voidage (ε_∞) are only function of Uo and Gs; they are determined according to a model by Patience and Chaouki (1993):

$$V_{p\infty} = \frac{Gs}{\rho_p\left(1-\varepsilon_\infty\right)};\ \varepsilon_\infty = \frac{Uo\rho_p}{\left(U_o\rho_p + \phi Gs\right)}; \qquad \phi = 1 + \frac{5.6}{Fr} + .47\,Fr_t^{.41} \tag{10}$$

The terminal velocity, U_t, was computed with a model proposed by Kunii and Levenspiel (1991). In this hydrodynamic model, the radial non-uniformity or the core-annulus flow structure are lumped into the parameter K, the fully developed velocity ($V_{p\infty}$) and the voidage (ε_{∞}.) By replacing K, the momentum Equation (6) becomes:

$$V_p \frac{dV_p}{dz} = g \frac{\rho_g - \rho_s}{\rho_s} \left[\left(\frac{U_o / \varepsilon - Vp}{U_o /_{\varepsilon\infty} - V_{p\infty}} \right)^{1.4} - 1 \right] \tag{11}$$

$$Uo = \left(\frac{W_{air}}{Mm_{air}} + \frac{W_{nat}}{Mm_{nat}} \right) \frac{RT_g}{P_{tot} A_r} \tag{12}$$

The superficial gas velocity, Uo, is computed by applying the ideal gas law and considering a constant volume reaction. The fluid properties are nearly those of air as both combustion gas and air contain over 70% of nitrogen, Table 2.2.

The voidage as a function of riser height, $\varepsilon(z)$ and the solid inventory in the riser, m_{sr}, are found from:

$$\rho_p \frac{d\varepsilon}{dz} = \frac{G_s}{V_p^2} \frac{dV_p}{dz} \tag{13}$$

$$\frac{d}{dz}(m_{sr}) = A_r \rho_p (1 - \varepsilon); \qquad m_{sr} = 0 \text{ at } z = 0 \tag{14}$$

$$\frac{d}{dz}(m_{sr}) = A_r \rho_p (1 - \varepsilon); \qquad m_{sr} = 0 \text{ at } z = 0 \tag{15}$$

Pressure drop is the sum of particle acceleration forces, gravitational forces, gas to tube frictional forces and particles to tube frictional forces. Including the frictional forces, one writes:

$$-\frac{dp}{dz}=\rho_p(1-\varepsilon)V_p\frac{dV_p}{dz}+\rho_p(1-\varepsilon)g+F_{g-w}+F_{s-w} \quad (16)$$

Table 2.2: Thermo-physical properties of fluid gas, sand particles and construction materials

Material	Mass kg	Heat Capacity C_{pw}, J/kg.K	Thermal Conductivity, k_w, W/m² K	Density, ρ_w, kg/m³	Emissivity ξ	Viscosity μ Pa.s
Fluid gas: $i \equiv O_2, H_2O, CO_2, N_2$		$C_{pg}=\sum y_i C_{pi}$	0.064	$P_{tot}M_m/RT$	$\xi=\xi(y_i,T)$	$\mu_g=\mu_g(T)$
Sand		840	1.7	2600	0.9	
Stainless steel-outer cylinder	642	502	15.14	7910	0.15	
Stainless steel-Riser	36	502	15.14	7910	0.15	
Refractory	480	920	1.8	2670	0.9	
Blanket	42	1130	0.01-0.37	128	0.9	
Total mass	1200					

*heat capacities of components from Moses (1978). emissivity from(Taylor and Foster. 1974).Viscosity. from Touloukian (1977).**from Moses(1978). other data are from the provider.

2.4.2.2 Annular bed

The annular bed is similar to the standpipe of CFB. It is assumed to be in plug flow with solids moving at a velocity proportional to Gs. This assumption was also verified by a thermal tracer analysis performed with the injection of cold sand into the hot annular bed. In Figure 2.4, thermal signals show three peaks of nearly identical standard deviation that confirms the assumption of piston flow. The voidage of the bed is equal to that of a fixed sand bed ($\varepsilon\approx0.476$). G_s measurements contain an experimental error of less than 10%.

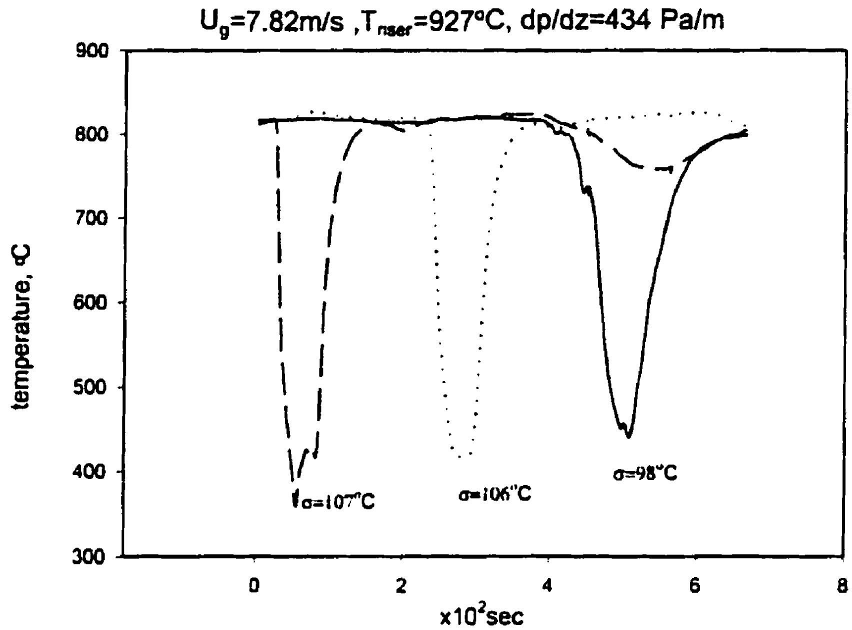

Figure 2.4: Thermal tracing in the annular sand bed obtained by injection of cold sand tracer signals monitored by three thermocouples located at different axial positions

2.4.2.3 Disengagement zone

Prior to the disengagement zone, gas and solid are completely separated by the impact device, which causes intense turbulence in the gas phase. This zone is modeled as a completely mixed zone (CSTR).

2.4.3 Mass Balance And Analysis

The following assumptions are made: i) Reaction occurs in the riser only; ii) Only oxygen diffuses to the particle surface where combustion occurs. Internal diffusion is ignored because sand particles are non-porous; iii) Reaction does not take place in the annulus where gas percolation is negligible; iv) Particle size is unchanged by reaction, as the coating resin constitutes less than 5% of the sand particle; v) Methane and air react completely in the burner. vi) Resin concentration and voidage vary with height only.

2.4.3.1 Riser

The unsteady state mass balance is written by considering the riser as a pseudo-plug flow reactor. For resin:

$$\rho_p A_r(1-\varepsilon)\frac{\partial Y_{pr}}{\partial t} = (W_s + W_{feed})\frac{\partial Y_{pr}}{\partial z} - \rho_p A_r(1-\varepsilon)k_r Y_{pr}^2 Y_{O2,S}^{1.3} \quad (17)$$

And for oxygen:

$$\frac{\rho_g}{M_{m,g}} A_r \varepsilon \frac{\partial Y_{O2}}{\partial t} = \frac{W_g}{M_{m,g}}\frac{\partial Y_{O2}}{\partial z} - \rho_p A_r(1-\varepsilon)k_{O2} Y_{pr}^2 Y_{O2,S}^{1.3} \quad (18)$$

Equation (17) and (18) contain the surface concentration of oxygen, $Y_{O2,S}$, which is linked to the bulk concentration by equating the oxygen diffusion rate to the reaction rate as :

$$N_{O2} = k_{mO2}\frac{P_{tot}}{RT}(Y_{O2} - Y_{O2,S}) = r_{O2} = k_{O2} Y_{O2,S}^{1.3} Y_{pr}^2 \quad (19)$$

The mass transfer coefficient, k_{mO2}, is computed according to a correlation by LaNauze et al. (1984):

$$Sh = \frac{k_{mO2} d_p}{D_{eff}} = 2\varepsilon + \left[\frac{4d_p(U_o/\varepsilon - V_p)}{\pi D_{eff}}\right]^{1/2} \quad (20)$$

The oxygen diffusivity, D_{eff}, is adopted from Vargaftik (1975):

$$D_{eff} = 1.0554 \times 10^{-9} T^{1.75} \tag{21}$$

At time t=0, the reactor contains no resin. At the riser base, z=0, is the location of the burner: complete combustion of natural gas is assumed, and the inlet resin concentration is the average of fresh feed resin and annular bed resin. With, Ws = G_s A_r, initial and boundary conditions are formulated as:

$$Y_{p,r}\Big|_{t=0} = 0; \qquad Y_{p,r}\Big|_{z=0} = \frac{W_{feed} Y_{p,feed} + W_s Y_{pa}\big|_{z=0}}{W_{feed} + W_s}; \; Y_{O2} = \left\langle \begin{matrix} \text{for natural - gas} \\ \text{complete combustion} \end{matrix} \right\rangle \tag{22}$$

2.4.3.2 Annular bed

With the assumption of plug flow and no reaction, the mass balance on resin is:

$$\rho_p A_a (1-\varepsilon_a) \frac{\partial Y_{pa}}{\partial t} = W_s \frac{\partial Y_{pa}}{\partial z} \tag{23}$$

Initial and boundary conditions are:

$$t = 0 \;,\; Y_{r,an} = 0 \qquad ; \qquad z = H_r \;(\text{riser exit})\; Y_{pr} = Y_{pa} \tag{24}$$

Concentrations in the riser and in the annular bed are interrelated through boundary conditions at the exit and the inlet of each zone (Equations (22), (23) and (24)).

2.4.4 Energy Balance And Analysis

The fluidized beds and the CFB in particular are well known for their temperature uniformity (Kunii and Levenspiel, 1991). Temperature values in the different zones of the reactor are presented in Figure 2.5 as a function of Gs. Because of its recent development, literature on thermal characteristics of ICFB is scarce. However, Milne et al. (1994) have published some data on temperature profile in an ICFB which show that axial and radial temperature gradients in the riser are not important. This is confirmed for the axial gradient in Figure 2.5. In the annular bed, radial gradients become evident only at the proximity of the inner walls of the refractory, Figure 2.5. The mass of solid has nearly the same and uniform temperature as the riser walls. Unsteady state energy equations are written for the gas and solid phases in each zone and the walls according to the following assumptions:

i)Radial and axial profiles of temperature in the riser are neglected due to the high mixing of gas-solid flow in the riser, and the presence of the annular sand bed which insulates the riser walls. ii)Uniform temperature within sand particles ($Bi \ll 1$) which are instantaneously heated up to the combustion temperature and absorb the heat generated by resin combustion. iii)Heat losses occur through the external cylinder walls made of refractory, mineral blanket and stainless steel. Thermal properties of these materials are listed in Table 2.2. All external walls of the cyclone system, the burner and the upper and lower plates are lumped into a single stainless steel cylinder, with the same volume of refractory and mineral blanket being uniformly redistributed inside, to result in a cylinder of equivalent hydraulic inside diameter $D_{w,i}$ and outside diameter $D_{w,o}$. This is acceptable as the vertical walls of the cylinder account for about 80% of the heat transfer area.

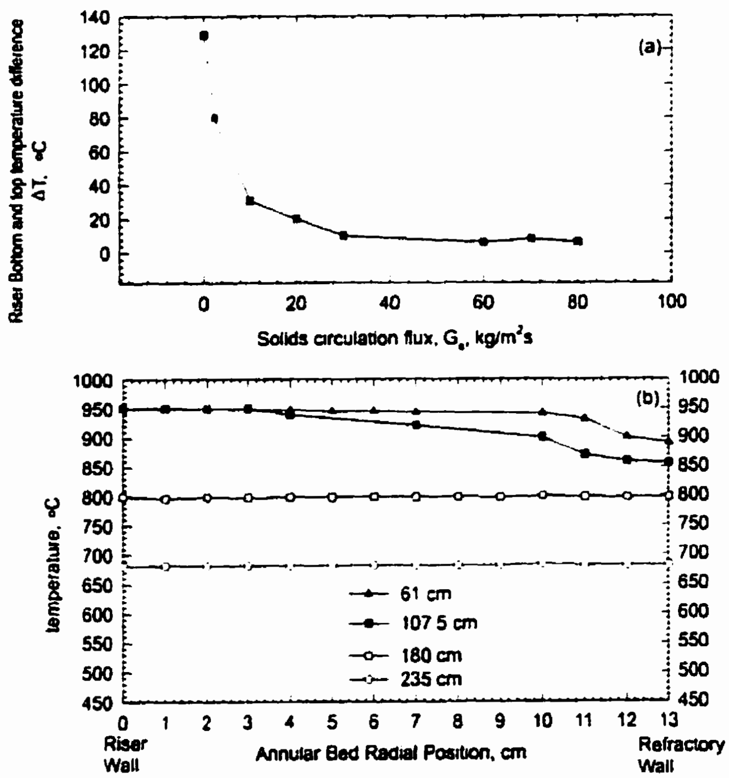

Figure 2.5: a--bottom and top temperature difference in the riser. b—Annular and disengagement zone radial temperature profiles at different axial positions.

2.4.4.1 Riser

Energy balance on solid particles:

$$m_{sr} C_{ps} \frac{dT_{pr}}{dt} = C_{ps}\left[W_s (T_{pa} - T_{pr}) + W_{feed}(T_{feed} - T_{pr})\right] + A_{g-p}[\sigma(\varepsilon_g T_{gr}^4 - T_{pr}^4) + h_{gp}(T_{gr} - T_{pr})] + (W_s + W_{feed})[Y_{pr}|_{z=0} - Y_{pr}|_{z=Hr}]\Delta H_{resin} \tag{25}$$

Energy balance on gas phase:

$$m_g C_{vg} \frac{dT_{gr}}{dt} = W_g C_{pg}(T_{feed} - T_{gr}) - A_{g-p}\left[\sigma(\xi_g T_{gr}^4 - \alpha_g T_{pr}^4) - h_{gp}(T_{gr} - T_{pr})\right] - A_{w-g}\left[\sigma(\xi_g T_{gr}^4 - \alpha_g T_{pa}^4) - h_{w-g}(T_{gr} - T_{pa})\right] + W_{nat}\Delta H_{nat} \tag{26}$$

Where, $C_{pg} = RM_{m,g} + C_{vg}$, is the temperature dependent molar average heat capacity of gas, and the emissivities are in Table 2.2. The mass of solid particles in the riser, m_{sr}, is computed from Equation (11). The gas-particle heat transfer coefficient, h_{pg}, and fluid to wall heat transfer coefficient, $h_{w\text{-}g}$, are taken from Rowe and Claxton (1965) and Sadek (1972) respectively.

Initial and boundary conditions: Solids from the annular bed and solids from the feeding hopper enter at the riser base. Gas and solids enter the bed at ambient temperatures, the initial temperature depend on the operation mode:

$$z = 0,\ T_{p,r} = \frac{W_{feed} T_{feed} + W_s T_{p,a}\big|_{z=0}}{W_{feed} + W_s}\ ;\ T_{g,r} = T_{amb} \tag{27}$$

2.4.4.2 Annular bed

The very high heat capacity solids undergo a small drop in temperature as they flow through the annular bed. This temperature drop is less than 20°C between the top and the bottom as compared to the driving force to the surroundings of 700~850°C. Ignoring annular gas percolation, the heat balance is written as:

$$m_{s,tot} C_{ps} \frac{dT_{pa}}{dt} = (W_s + W_{feed}) C_{ps} (T_{pr} - T_{pa}) - Q_{loss1} \tag{28}$$

$$Q_{loss1} = \pi D_{w1} H_a \left[h_{pw} \left(T_{pa} - T_w\big|_{r=r1} \right) + F_w\, \xi_w\, \xi_p \sigma \left(T_{pa}^4 - T_w^4\big|_{r=r1} \right) \frac{H_c}{H_a} \right] \tag{29}$$

The convective heat transfer coefficient between the particles and the refractory, h_{pw}, is computed from a correlation by Colakyan and Levenspiel (1984) for a moving bed. The view factor, F_w, is unity.

Initial and boundary conditions are similar to those for mass balance equation:

$$T_{p,a}\Big|_{z=H_a} = T_{p,r}\Big|_{z=H_a} \quad ; \quad T_{p,r}\Big|_{z=0}(W_s + W_{feed}) = W_s T_{p,a}\Big|_{z=0} + W_{feed} T_{feed} \tag{30}$$

2.4.4.3 Disengagement zone

The disengagement zone contains no solids. It is modeled as a CSTR reactor:

$$m_{g,f} C_{v,g} \frac{dT_{g,f}}{dt} = W_g \int_{T_{pr}}^{T_{g,f}} C_{p,g} dT - Q_{loss2} \tag{31}$$

$$Q_{loss2} = \pi D_{w,i}(H_c - H_a)\left[\sigma \xi_g \, \xi_w \left(T_{g,f}^4 - T_w^4\Big|_{r=ri}\right) + h_{gw}\left(T_{g,f} - T_w\Big|_{r=ri}\right)\right] \tag{32}$$

The heat transfer coefficient, h_{gw}, is from the correlation for gas flow in pipes (Holman, 1997).

2.4.4.4 Heat Losses

The overall heat losses are computed by considering the wall as a single material with radially changing thermal properties. Contact resistances between adjacent materials are ignored.

$$\rho_w C_{pw} \frac{\partial T_w}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(k_w r \frac{\partial T_w}{\partial r}\right) \tag{33}$$

and boundary conditions:

$$r = ri = \frac{D_{w,i}}{2}, \qquad Q_{loss1} + Q_{loss2} = \pi D_{w,i} H_c \left(-k_{w,i} \frac{\partial T_w}{\partial r} \right) \tag{34}$$

$$r = ro = \frac{D_{w,o}}{2}, \qquad A_{w,o}\, h_{w,o} \left(T_w - T_{amb} \right) = \pi D_{w,o} H_c \left(-k_{w,o} \frac{\partial T_w}{\partial r} \right) \tag{35}$$

$A_{w,o}$ includes all the external area of cylinder and its flanges. The external heat transfer coefficient, $h_{w,o}$,is consistent with heat loss measurements on the same unit. Heat losses are about 15~20% of total energy input.

2.5 Simulation Results

The simulation is performed to predict the transient heating of clean sand with natural gas and the dynamic behavior during treatment of foundry sand resin. The initial condition depends on flowrate of natural gas and air, and on the hydrodynamic conditions (G_s and solid loading). The model simulates the process at high temperature. Simulation of waste treatment starts after steady state heating of clean sand is reached.

2.5.1 Computer Modeling Strategies

The transient and steady state equations are solved numerically with routines combining Petzold-Gear BDF method (Brenan et al., 1989), Runge-Kutta method, modified Powell Hybrid method (Powell, 1970) and finite difference methods. The finite difference methods transform the "d/dz" into algebraic form. The Runge-Kutta method is used to compute the hydrodynamics. These methods are combined to Petzold-Gear BDF method and Powell Hybrid method to predict the transient and the steady state respectively.

2.5.2 Hydrodynamic predictions

The proposed modeling allows the study of the pilot-scale reactor behavior under thermal treatment conditions. The hydrodynamic model allows the prediction of the pressure drop, the voidage, the solids loading, the residence time and the particle velocity in the riser. Model predictions for these parameters are presented in Figure 2.6. The prediction of pressure drop in the riser is shown in Figure 6a along with experimental data, for mass fluxes and gas velocity ranging from 20 to 120 $kg/m^2.s$ and from 7.8 to 11.4 m/s respectively. Overall, the predictions show good agreement with experimental data which are in the same range as other published data by Patience (1992), Pugsley and Berruti (1996) , and Nieuwland et al. (1997) that is between 200 and 1000 Pa/m. The solids circulation flux has much more influence on the pressure drop than the superficial gas velocity. This is because Gs has a strong effect on the voidage (see Figure 2.6c) and on the acceleration of particles (see Figure 2.6d) and gas friction is negligible for dilute gas-solid flow. The calculated voidage is indirectly justified by pressure drop data. The model calculates residence times of the order of 0.5 to 1.3 s (see Figure 2.6b). The calculations show that the residence time is directly proportional to solids loading in the riser and to a greater extent, inversely proportional to the superficial gas velocity. The solid velocity shown in Figure (2.6d) is the sectional average value at any axial position, not the velocity of a single particle. Therefore, depending on the value of the suspension gas velocity, the acceleration zone could extend from 1.0 m to 3.0 m. Therefore, in the present ICFB riser, with a length of 1.5 m, mostly operates within the acceleration zone.

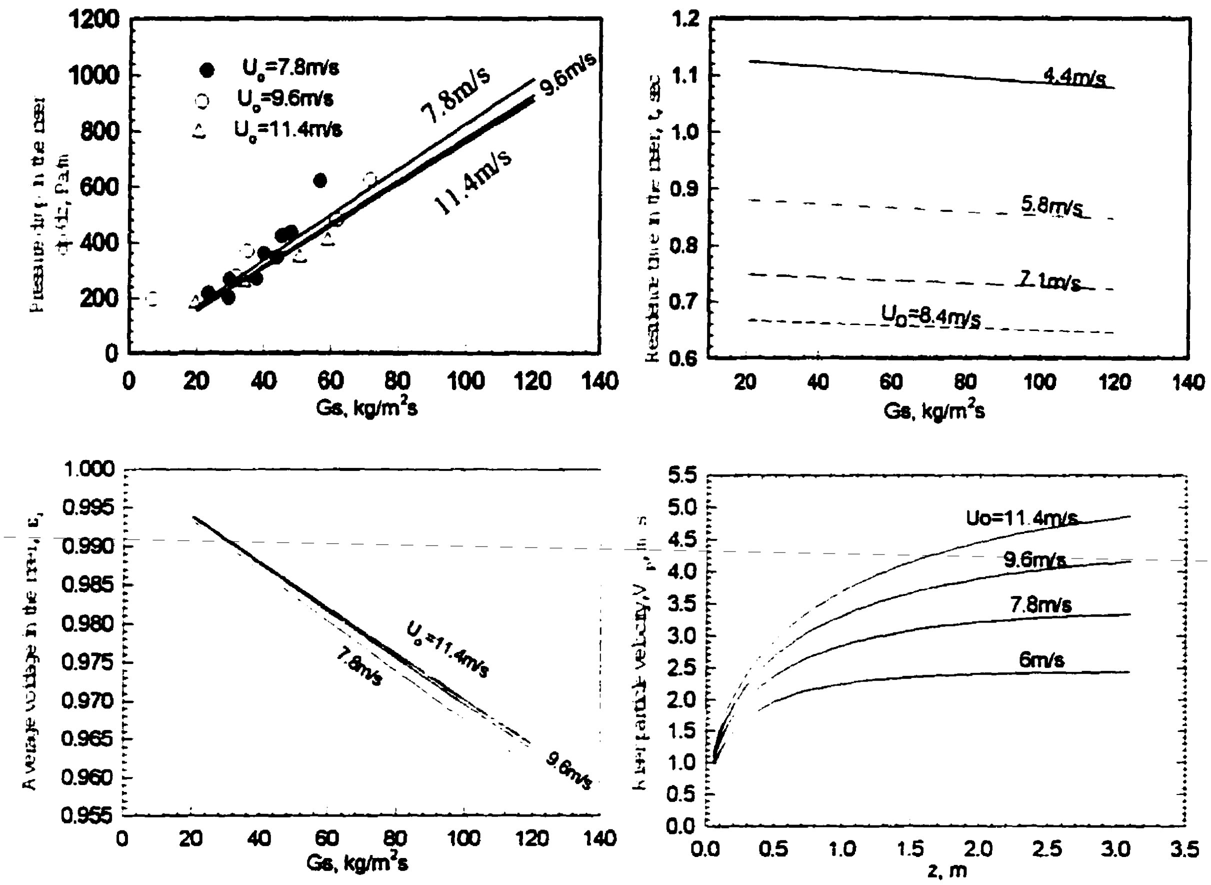

Figure 2.6: Hydrodynamic properties in the riser: computed and experimental data; a- pressure drop in the riser; b- average particle residence time in the riser; c- average voidage in the riser; d- sectional average velocity of particles in the riser. Temperature range: 850 to 1000°C.

2.5.3 Prediction of temperatures and resin removal

The calculation was performed with thermo-physical properties listed in Table 2.2. The model predicts the steady and unsteady state behavior of the reactor, with and without resin combustion taking place. In Figure 2.7, the computed transition temperature to steady state (clean sand, no resin) show good agreement with experimental data in the riser and the annular bed. Steady state is reached after 15 to 20 hrs of heating the bed. At t=0, as gas flows into the riser, an initial sharp rise in temperature is compensated by the circulation of cold solids (>600 kg/h) which continuously lose energy through the refractory walls. In Figure 2.8, one sees that steady state temperatures are lower for continuous feed of solids with resin. This is because the calorific contribution of foundry sand resin is not high enough to overcome the cooling effect created by feeding cold material into the reactor. Although the resin has a heating value of about 26 MJ/kg, its low concentration (≈2%) in the foundry sand results in an overall waste of heating value less than 1MJ/kg which is below the self-combustion limit of about 2.3MJ/kg (Brunner, 1991). According to the operating feedrates, the calorific contribution of resin is less than 20%. The transient temperature curves in Figure 2.8 show that the bed with smaller solids inventory has less thermal inertia to resist thermal perturbations; as observed, an initial cooling period is followed by a heat-up period when foundry sand is fed into the bed with smaller solids inventory. Feeding of clean sand without resin would simply produce a cooling effect, as no additional resin combustion would be involved and the steady state temperature would be lower. Figure 2.8 also compares measured and calculated resin conversion for operating times of up to 7 hours for solid feedrate of 36 and 41kg/h. The results show that the model is able to predict the resin conversion within experimental errors. With combustion of foundry sand, steady state operation is reached much earlier than the bed replacement process which would require approximately 7 to 12 hours of operation; as shown in Figure 2.8, the model can correctly predict the transient behavior of the bed.

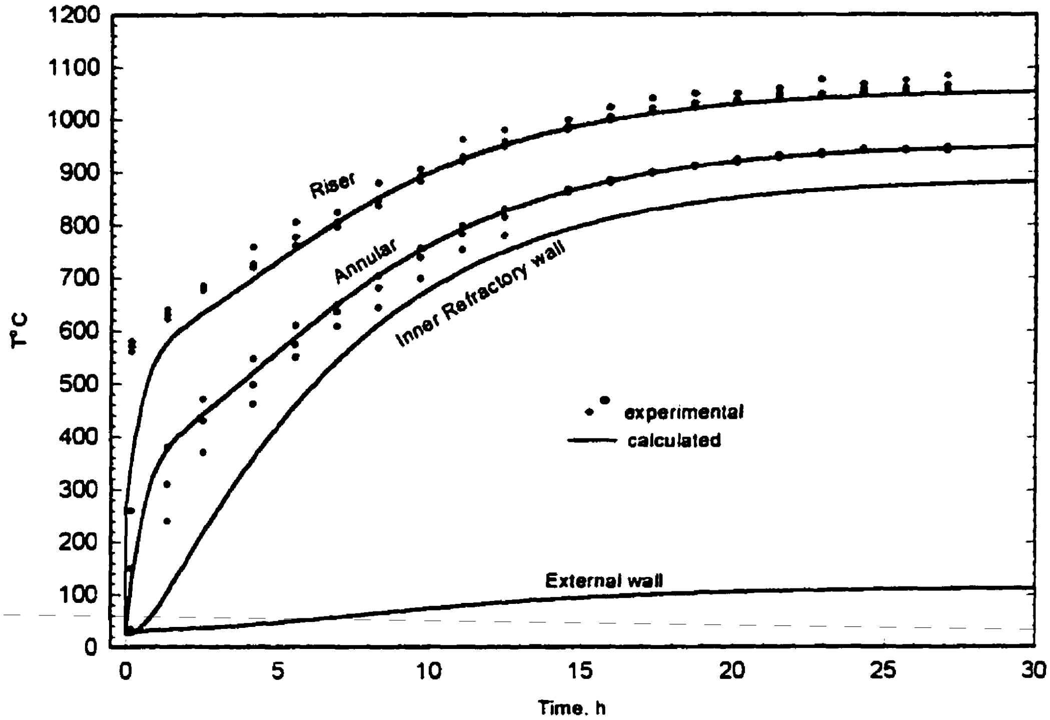

Figure 2.7: Comparison between model and experiment for the transient temperature in the ICFB during the clean sand heating period from room T=35°C, $m_{s,tot}$=140 kg, W_{air}=37.7 kg/h, aeration=1.4, Gs=35 kg/m^{2}s, ws=0 kg/h, $k_{blanket}$ = k(T) W/m°C, k_{sand}=1.82 W/m°C, $h_{w,o}$=13 W/m^2 °C

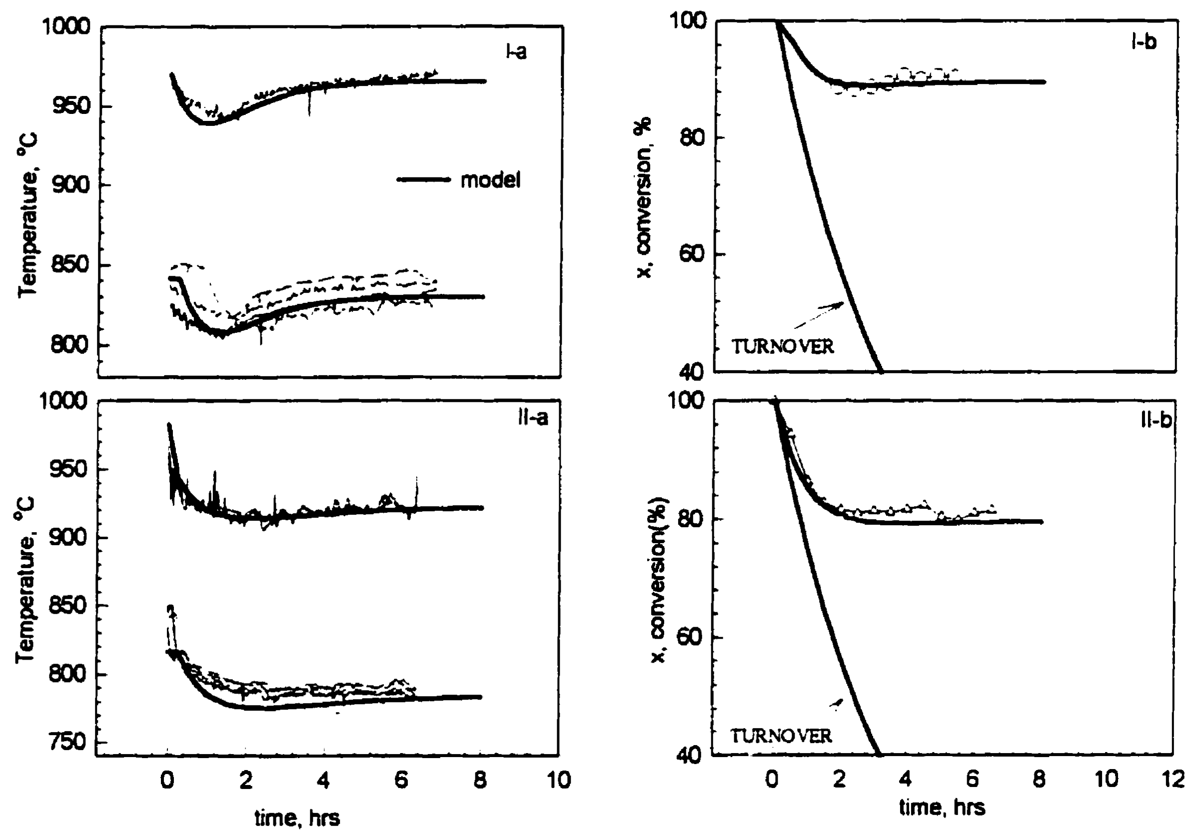

Figure 2.8: Comparison between model and experimental transient temperatures (I-a, II-a) and conversions (I-b, II-b) during treatment of foundry sand. I-a,b: W_{air}=37.7 kg/h, aeration=1.35, W_{feed}=35 kg/h, $m_{s,tot}$=100 kg, Gs=35 kg/m^{2}s, from room T=35°C, $h_{w,o}$=13 W/m^2 °C, $h_{w,i}$=32 W/m^2 °C, $k_{blanket}$=k(T) W/m°C. II-a,b: W_{air}=34.8 kg/h, aeration=1.2, W_{feed}=41 kg/h, $m_{s,tot}$=140 kg, Gs=50 kg/m^{2}s, from room T=15°C, $h_{w,o}$=13 W/m^2°C, $h_{w,i}$=33 W/m^2°C, $k_{blanket}$=k(T) W/m°C.

Figure 2.9 shows the effect of solids circulation flux (G_s) on the conversion for the riser and the entire reactor. It suggests that for the same solid feedrate, higher G_s yields higher overall conversion despite having lower single pass riser conversion. This is because the solids spend more time into the riser (the site of reaction) at higher G_s as the residence time of solids in the annular bed is shorter and the number of cycles is greater. By defining the cycle time as the sum of particles residence time in the riser and residence time in the annular bed:

$$t_{cycle} = t_{riser} + t_{annular} \text{ ; with } t_{annular} = \frac{m_{s,tot}}{G_s A_r} \tag{36}$$

The overall residence time is:

$$\tau = \frac{m_{s,tot}}{W_{feed}} \tag{37}$$

At a fixed feedrate of solids, the number of cycles through the riser is calculated as the ratio τ/t_{cyc}. Because $t_{an} >> t_{riser}$, reduction of annular residence time due to an increase in solids circulation will generate an increase of the number cycles through the riser but also a shorter residence time for each cycle. The overall effect is an increase in the conversion of the waste because of an increase in the global riser residence time, although it is diminished for each cycle. Figure 2.9 reveals also that axial concentration gradients and single pass riser conversions are lower at higher G_s because of increased backmixing effect. The solids circulation flux, G_s, acts as a mixer, and at high ratio of Gs to feedrate, the reactor resembles more a CSTR. In practice, G_s can not be increased indefinitely because solids-flame contact can only exist in dilute gas-solid flow. Highly dense flow would provoke flame quenching or extinction. The predicted axial conversion of resin in the riser exhibits a nearly linear increase with riser height. Thus by increasing the riser height, one can still expect significant gain in conversion provided that temperature remains constant. This results is encouraging as it indicates that the ICFB holds great promise as a reactor for treatment of industrial waste and for fast, competing reactions such as thermal pyrolysis and gasification where complete combustion of hydrocarbons is not desired. The residence time of solids in the riser being of the order of 1s (Figure 2.6b) suggests that the ICFB can be used for reactions that require short contact times. Figure 2.9 indicates that solid circulation flux, G_s, and riser height are the two major design parameters for the reactor.

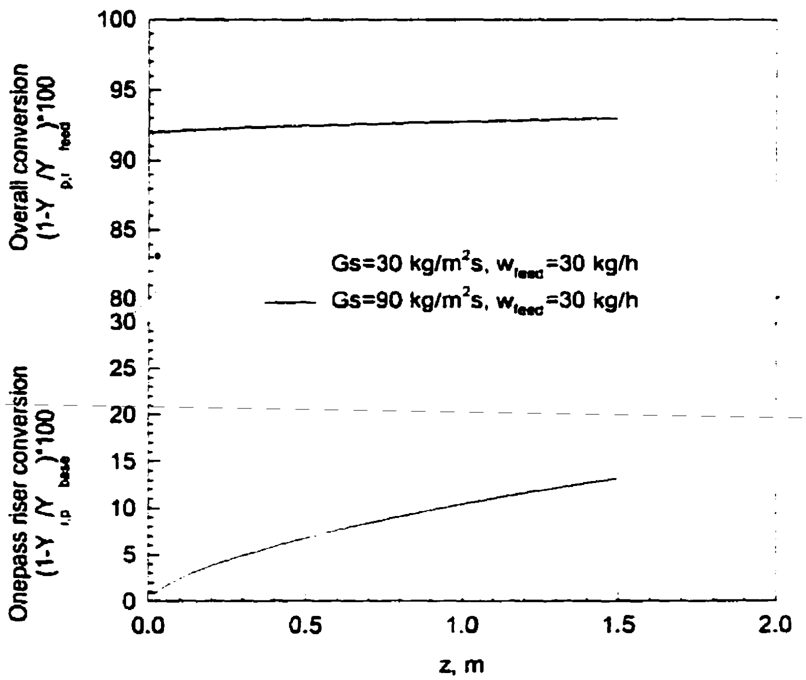

Figure 2.9: Model prediction of the effect of solid circulation on the treatment of spent foundry sand. W_{air}=37.7 kg/h, aeration=1.4, $m_{s,tot}$=140 kg, $h_{w,o}$=13 $W/m^2\,°C$.

2.6 Conclusion

A predictive model is developed to describe and characterize all key design variables in a novel Internally Circulating Fluidized Bed Combustor. The model being based on fundamental principles as well as some empirical correlations, is suited to study both the transient and steady state behavior of an ICFB reactor.

The input parameters to the model are: gas and solid waste flowrates, circulation flux, reactor geometry, and physical properties of gas and solids. The simulation allows the prediction of residence time, voidage, pressure drop across the riser, particle velocity, temperatures as well as waste conversion. Comparison of model predictions with experimental data during thermal treatment of spent foundry sand shows good agreement.

The predictions suggest that the Internally Circulating Fluidized Bed holds promise as a reactor for thermal treatment of organic material and waste. The presence of the riser is suitable to confine the high temperature region while keeping the rest of the reactor at relatively lower temperature, hence increasing its thermal efficiency. The study also yields that the feedrate and the solid circulation flux can be manipulated to produce desired reactor performance.

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Chapitre 3___PREDICTION OF GAS EMISSIONS IN AN INTERNALLY CIRCULATING FLUIDIZED BED COMBUSTOR FOR TREATMENT OF INDUSTRIAL SOLID WASTES

Reference: L. Mukadi, C. Guy and R. Legros(1998): " Prediction of Gas Emissions in an Internally Circulating Fluidized Bed Combustor For Treatment of Industrial Solid Wastes ", Submitted to Fuel.

Keywords: Fluidized bed combustion, simulation, pollutant gas emission, wastes.

3.1 Contexte[a]

Dans le chapitre précédent, un modèle mathématique a été développé pour prédire la conversion, la température, le temps de séjour, la porosité et la vitesse des particules dans le tube de montée. Le modèle est basé sur des hypothèses et des équations mathématiques relatives à l'hydrodynamique, aux transferts de chaleur et matière et la cinétique de réaction. Le modèle pour le transfert de chaleur a été d'abord développé puis validé en utilisant des coefficients de transfert de chaleur appropriés. Ce modèle a été ensuite combiné à la cinétique de réaction pour caractériser la performance du réacteur représentée par la conversion ou la destruction du déchet solide.

Les autres paramètres indicatifs de la performance environnementale du procédé doivent être maintenant déterminés. C'est l'objet du présent chapitre qui propose une approche conduisant à la prédiction des émissions gazeuses produits lors du traitement thermique de déchets. Ainsi, le système de réaction est maintenant plus complexe pour inclure des réactions homogènes et hétérogènes importantes lors de la formation et la réduction des contaminants gazeux dans les fours à lits fluidisés.

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[a] ne pas inclut dans l'article

3.2 Abstract

Simulations of an ICFB waste combustor are reported with a mathematical model which includes reaction kinetics related to carbon, nitrogen, hydrogen, oxygen and sulfur content of industrial waste. The model assumes formation of volatile and char during the thermal treatment of wastes, followed by a series of homogeneous and heterogeneous reactions catalyzed by bed material such as char and ash+sand. The trends of model predictions are in qualitative agreement with experimental observations from thermal treatment of spent foundry sand in a 20 kW ICFB pilot unit. According to the kinetics and experimental data, bed material are strongly responsible for low level emissions of nitrogen oxides and sulfur dioxide. The validated model allows a comprehensive simulation analysis to be performed by varying some operating parameters such as waste feedrate, aeration rate, solid circulation and reacting zones. The study reveal that for proper combustor operation, a balance has to be made between combustion efficiency, lower carbon monoxide and nitrogen oxides emission, as well as sulfur dioxide capture

3.3 Introduction

Fluidized bed combustion is increasingly accepted for disposal, recycling and energy recovery from industrial wastes. This process generates flue gases whose composition depends on properties of the wastes and operating conditions of the combustor. In general, fluidized bed combustion is a clean technology for combustion of solid fuels, biomass and wastes. However, the increasingly stricter environmental regulation requires even lower emissions.

For industrial wastes of organic origin, gas emissions of most concern are CO, CO_2, NOx, N_2O, SO_2, O_2 and the unburned hydrocarbon. These products are well-known pollutants causing photochemical smog, acid rain and global warming due to greenhouse effect and ozone depletion; some of them have also adverse effects on human health. Several studies show that gaseous emissions from fluidized bed combustors are the result of homogeneous and heterogeneous formation coupled with *in situ* destruction reactions [Li, 1998; Anthony, 1995; Boavida et al., 1997; Desai et al.,1995; Goel et al. 1994; Goel et al., 1996; Jenssen and Johnsson 1997; Philippek et al., 1997; Talukdar and Basu, 1995, Winter et al., 1996 and 1997]. Reactions related to such products are complex and not fully understood as intermediate products play a major role in the overall reactions and many of the homogeneous and heterogeneous reactions are catalyzed by solid material in the combustor. The hydrodynamics also has an influence on the gaseous emissions. Comprehensive modeling may help to obtain a better understanding of the process and to guide the optimization procedure towards lower pollutants emission levels. Several modeling efforts have been made to predict gaseous emissions from fluidized bed combustion of organic materials such as coal, char, biomass and waste. [Hannes et al., 1995, 1997; Jenssen and Johnsson, 1997; Tsuo, 1995; Borodulya et al.1995; Agrawal and LaNauze,1989; Talukdar and Basu, 1995; Johnsson, 1989; Lin and Bleek,1991, Merrick, 1983; Reddy and Mohapatra, 1994]. As there are no published kinetics for emissions in the fast fluidized bed regime, one may apply all the available kinetics models related to carbon, nitrogen, sulfur, hydrogen and oxygen to predict flue gas concentration in the combustor provided that the composition of gas and solids inside the combustor are known, which is a complex and difficult task. However, the work of Merrick (1983), Johnsson (1989), Lin and Bleek (1991), Talukdar and Basu (1995), Jensen and Johnsson (1997), Goel et al. (1996), Philippek et al. (1997) suggest that only some reactions are important for the formation of pollutant emissions in fluidized bed. Hence, a simplified model is appropriate for a rapid estimation of the pollutant concentrations and process analysis. Such a model could serve effectively for design and operation purposes, to predict the effect of different design parameters, operating conditions and the influence of waste properties on the pollutant emissions.

The objective of this work is to present a simple mathematical model that comprehensively describes the related chemical reactions, and reasonably predicts CO, CO_2, NOx, N_2O SO_2, O_2 concentrations from flue gas emissions of a novel Internally Circulating Fluidized Bed (ICFB) combustor developed by Guy et al.(1997). The ICFB experimental unit presented in Figure (3.1) is made of two concentric cylinders, which form the riser and the annular bed region, a cyclone and a gas and solids feed system. A screw regulates feedrate of fresh solids which are transported pneumatically into the riser. Fresh solids and solids from the annular bed mix at the riser base, and are carried up by the flowing gas. At the riser top, the presence of an impact device and the disengagement zone provide gas-solids separation. The gas and a small amount of fine particles flow out to the cyclone, while the separated solids move downwards through the annular bed to be re-injected at the base of the riser. Measurements of temperatures, pressures and solid withdrawals are performed at different axial positions in the reactor. Type B thermocouples with ceramic shields are used to measure temperatures in the riser. Type K thermocouples are used for all other temperature measurements. Concentrations of NOx, N_2O, CO, CO_2, SO_2, O_2 and unburned hydrocarbons are obtained from on-line gas analyzers.

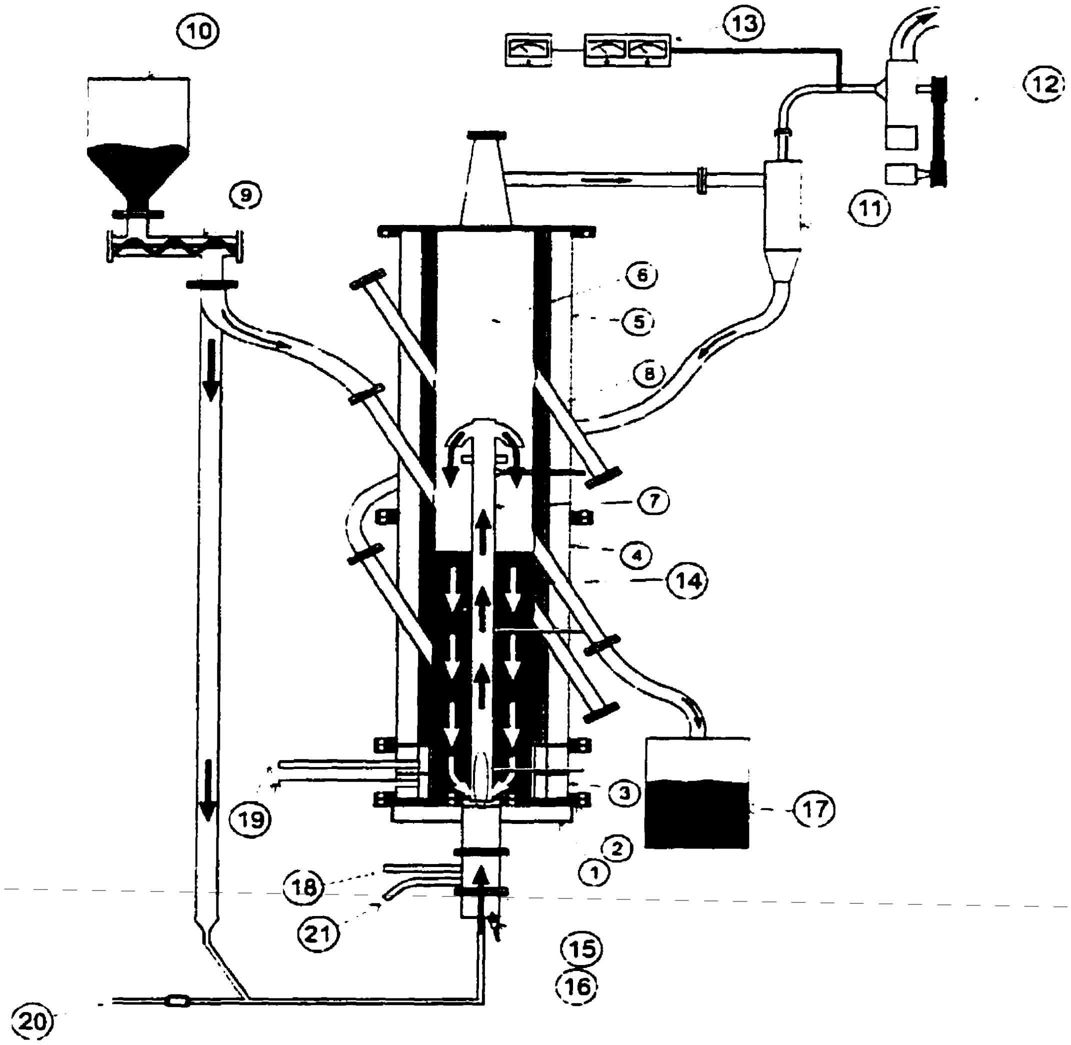

Figure 3.1: Schematics of the Internally Circulating Fluidized Bed pilot unit: 1-Plenum, 2-Support plate, 3- Secondary air distributor, 4-Lower chamber, 5-Upper chamber, 6-Disengagement zone, 7-Riser, 8-Impact device, 9-Screw-conveyor, 10-Hopper, 11-Cyclone, 12-Ventilator, 13-Flue gas analyzer, 14-Annulus region, 15-Burner, 16-Spark plug, 17-Treated solids, 18-Primary air, 19-Secondary air, 20-Air for pneumatic transport, 21-Natural gas. Riser –length: 1.5m/diameter: i.d.780mm, o.d. 890mm. External cylinder– length: 2.5m/ diameter: i.d. 355mm, o.d.645mm. Burner length: 300mm

3.4 Model Development

The mathematical model accounting for heat and mass transfer in the ICFB combustor is presented in Mukadi et al.(1998). Here, the same model is applied with some additional assumptions and modifications specific to gaseous emissions during combustion of low heating value industrial wastes containing organic material.

3.4.1 Model Assumptions

1. All reactions take place in the riser, the disengagement zone and the cyclone. There is no reaction in the annular bed. Homogeneous reactions occur in the disengagement zone and the cyclone as the voidage is close to unity.

2. The wastes first convert to volatiles and char. The volatile are NH_3, CO, SO_2, H_2O alone. The char-(C, N, O, S, H) converts to CO, NO, N_2O, SO_2 and H_2O at a rate proportional to char combustion. Both heterogeneous and homogeneous reactions occur, as shown in Table (3.1).

3. A gas-solids core-annulus flow model is considered for the riser, assuming a core of radius, r_c, with plug flow of phases, gas stagnant in the annulus and an interchange coefficient, Kg, between the core and the annulus, gas is stagnant in the annulus, Figure(3.2). The disengagement zone and the cyclone are well mixed.

4. Radial gradients are ignored.

5. Diffusion of oxygen toward the solid waste is taken into account.

6. Heat and mass balances are developed as in Mukadi et al. (1998).

7. Combustion of supplemental natural gas proceeds to completion.

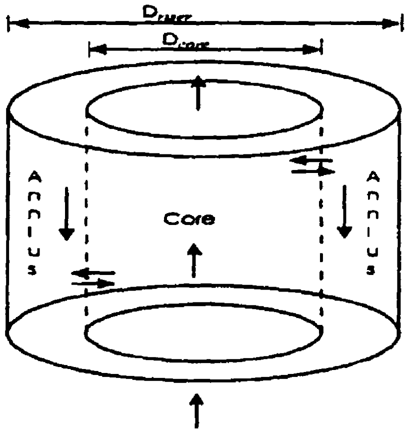

Figure 3.2: The core-annulus flow structure of the riser

3.4.2 Mathematical equations

Mass balance for each species inside the riser is written as:

$$\left.\begin{array}{l} -\dfrac{1}{\pi r_c^2}\dfrac{\partial F_{C,i}}{\partial z} - 2\dfrac{K_g}{r_c}\left(C_{c,i} - C_{a,i}\right) + \sum_i R_i = \dfrac{\partial C_{C,i}}{\partial t} \\ -2\dfrac{r_c}{R^2 - r_c^2}K_g\left(C_{a,i} - C_{c,i}\right) + \sum_i R_i = \dfrac{\partial C_{a,i}}{\partial t} \end{array}\right\} ; \quad \begin{array}{l} F_{C,i} = v\,C_{C,i} \\ Pv = \sum_i F_{C,i}\,R_g T \\ Y_{C,i} = F_{C,i} / \sum_i F_{C,i} \end{array} \tag{1}$$

with i≡components : CO, NO, SO_2, CO_2, O_2 , H_2O, N_2

Boundary conditions assume complete combustion of natural gas in the burner located at the riser base, with the natural gas being composed of methane only. The cross flow interchange coefficient, K_g, is obtained from the work of Patience and Chaouki (1993) :

$$\frac{K_g D\phi^{1/2}}{D_v \rho} = 0.25\left(\frac{\mu}{D_v \rho}\right)^{1/2}\left(\frac{\rho U_g D}{\mu \phi^{1/2}}\right)^{3/4}\left(\frac{G_s}{\rho_p U_g}\right)^{1/4} \tag{2}$$

The respective axial voidage of annulus and core are derived from the work of Zhang et al (1991):

$$\varepsilon_c = \int_0^{\phi_c} \phi^2 \varepsilon_{avg}^{\left(0.191+\phi^{2.5}+3\phi^{11}\right)} \qquad \phi = r/R,\ \phi_c = r_c/R \tag{3}$$

The average voidage in the riser, ε_{avg}, varies with height. It is computed using the hydrodynamic approach proposed in Mukadi et al. (1998). The ratio of core radius, ϕ_c, is computed according to a correlation by Patience and Chaouki (1993):

$$\phi_c^2 = \left[1 + 1.1Fr\left(\frac{G_s}{\rho_p U_g}\right)^{0.083Fr}\right]^{-1} \quad ; \qquad Fr = \frac{U_g}{\sqrt{gD}} \tag{4}$$

Equations in the disengagement zone and the cyclone are expressed as:

$$(vC)_{in,i} - (vC)_{out,i} = R_i V_{zone} \tag{5}$$

3.4.3 Related reactions and kinetics

An extended literature review yielded a number of reactions related to carbon, nitrogen, sulfur, hydrogen and oxygen. Table (3.1) and Table (3.2) present a generalized and simplified reaction scheme and the related kinetic rates. The scheme contains reactions which are considered to be relevant in the present case of organic waste as it accounts for heterogeneous and homogeneous reactions. The system includes the most important reactions identified in the literature for combustion of organic material in fluidized beds

[Merrick, 1983; Johnsson, 1989; Johnsson et al., 1991; Jensen and Johnsson, 1997; Johnson and Dam-Johansen, 1995; Johnsson et al., 1997; Winter et al., 1996; Philippek et al., 1997; Goel et al., 1996 Goel et al., 1994; Krammer and Sarofim, 1994]. The first step begins with the formation of volatile (as CO, NH_3, SO_2, H_2O) and char-(C,N,H,O,S), followed by a set of homogeneous and heterogeneous reactions. Less volatile wastes such as foundry sand may skip the first step of devolatilization.

Table 3.1: System of important reaction kinetics needed for model predictions

<table>
<tr><td colspan="2">Destruction of waste:
Waste → Volatiles-(CO, SO_2, NH_3, H_2O) → CO, NO, N_2O, SO_2, H_2O
Waste → Char--(C, H, N, O, S) → CO, NO, N_2O, SO_2, H_2O</td></tr>
<tr><td>Formation of CO and CO_2:
1. $\text{Char}-C + \frac{1}{2}O_2 \longrightarrow CO$
2. $CO + \frac{1}{2}O_2 \longrightarrow CO_2$

Formation and destruction of NO:
3. $2NH_3 + \frac{5}{4}O_2 \xrightarrow[\text{bed material}]{\text{Homo}} NO + \frac{3}{2}H_2O$
4. $\frac{1}{2}N_2 + \frac{1}{2}O_2 \rightarrow NO$
5. $\text{Char}-N + \frac{1}{2}O_2 \longrightarrow NO$
6. $NO + \text{char}-C \longrightarrow 0.5N_2 + CO$
7. $NO + CO \xrightarrow{\text{char, CaO}} \frac{1}{2}N_2 + CO_2$</td><td>Formation and destruction of N_2O :
8. $\text{Char}-N + NO \longrightarrow N_2O$
9. $N_2O \xrightarrow[\text{Bed-material}]{\text{homo}} N_2 + \frac{1}{2}O_2$
10. $N_2O + \text{char}-C \longrightarrow CO + N_2$
11. $N_2O + CO \xrightarrow[\text{Bed-material}]{\text{Char}} CO_2 + N_2$

Formation and capture of SO_2:
12. $\text{Char-S} + O_2 \rightarrow SO_2$
13. $CaO + SO_2 + 1/2\,O_2 \rightarrow CaSO_4$
Oxidation of ammonia to N_2:
14. $2NH_3 + 1.5O_2 \xrightarrow[\text{Bed-material}]{\text{Char}} N_2 + 3H_2O$</td></tr>
</table>

Table 3.2: Kinetic rates corresponding to reactions in table (3.1).

Reaction Rate(#), mol/s/m^3

1. $R_1 = F_C\, R_{CHAR} - R_6 - R_{10}$

2. $R_2 = k_2\, C_{CO}\, C_{H_2O}^{0.5}\, C_{O_2}^{0.25}\ \varepsilon$

   $k_2 = 1.78 \times 10^{10} \exp\left(\dfrac{21650}{T}\right)$

3. $R_3 = k_{3,homo} C_{NH_3} C_O$;

   $R_3 = k_{3,CATr} C_{NH_3} C_O C_{CAT} (1-\varepsilon)$

   $k_{3,homo} = 2.3 \times 10^{31} T \exp(-60000/T)$

   $k_{3,CaO} = 2.2 \times 10^{13} \exp(-9792/T)$

   $k_{3,char} = 1.268 \times 10^{16} \exp(-14752/T)$

4. $R_1 = k_1 C_{O2}^{1/2}\, C_{N_2}\varepsilon$

   $k_4 = 1.44 \times 10^{20} \left(\dfrac{1}{T}\right)^{1/2} \exp\left(-\dfrac{69460}{T}\right)$

5. $R_5 = \dfrac{F_N R_{Char}}{1 + k_{NO/N_2O} C_{NO}}$

   $k_{NO/N_2O} = 9.0 \times 10^2 \exp\left(-\dfrac{3551}{T}\right)$

6. $R_6 = k_6\, C_{NO}\, \varepsilon$

   $k_6 = 3.68 \times 10^8 \exp(-29700/T)$

7. $R_7 = k_{7,CAT}\, C_{NO}\, C_{CO}\ C_{CAT} (1-\varepsilon)$

   $k_{7,char} = 3.68 \times 10^7 \exp(-13097/T)$

   $k_{7,CaO} = 2100 T \exp(-8920/T)$

8. $R_8 = \dfrac{F_N k_{NO/N_2O} C_{NO}}{1 + k_{NO/N_2O} C_{NO}} R_{Char}$

9. $R_9 = k_{9,homo} C_{N_2O} \varepsilon$

   $R_9 = k_{9,CAT} C_{N_2O}\ C_{CAT} (1-\varepsilon)$

   $k_{9,homo} = 5.2 \times 10^9 \exp(-27000/T)$

   $k_{9,char} = 43.5 \times d_p^{-0.74} \exp(-1000/T)$

   $k_{9,Bed-material} = 1.7 \times 10^6 \exp(-22150/T)$

   $k_{9,CaO} = 1.3 \times 10^6 \exp(-13330/T)$

10. $R_{10} = k_{10} C_{N_2O}\ C_{CHAR} (1-\varepsilon)$

    $k_{10} = 4.8 \times 10^8 \exp(-16983/T)$

11. $R_{11} = k_{11,CAT}\, C_{N_2O}\, C_{CO}$

12. $R_{12} = F_S\, R_{char}$

13. $R_{13} = k_{13} C_{SO_2} C_{O_2} C_{CAO} (1-\varepsilon)$

14. $R_{14} = k_{14,CAT} C_{NH_3} C_O\ C_{CAT} (1-\varepsilon)$

    $k_{14,Char} = 0.6342 \times 10^{16} \exp\left(-\dfrac{14752}{T}\right)$

    $k_{14,CaO} = 2.663 \times 10^{12} \exp\left(-\dfrac{9755}{T}\right)$

Where C_{CAT} is the volume fraction of the catalytic material within the bed.

(#) rate 2 from Dryer and Glassman(1972), rate 3, 6, 7, 14 from from Johnsson (1989) and Johnsson et al. (1991) rate 4 from Zeldovich mechanism, in Kenbar et al. (1995); rate 5-8, 10 from Goel et al. (1996); rate 9 from Glarborg et al. (1994), Johnson and Dam-Johnson (1995), Johnsson et al. (1997); rate 11 from Philippek et al. (1997) rate 13 from Rajan and Wen (1980), Wen and Ishida (1973) and Borgwardt (1970).

The nitrogen content of the waste is transformed to NOx, N_2O and NH_3. The NOx is a mixture of NO and NO_2 where NO constitutes more than 90%. Hence, NO is used to represent NOx in equations of Table(1). This is sufficient as the two products are directly linked by formation of NO_2 at lower temperature after the stack. There are three principal sources of NOx: —thermal NOx, produced by Zeldovich mechanism, —the prompt NOx, produced in flame from the fuel rich flames, —the fuel-NOx, resulting from the nitrogen contained in the fuel. For the case where oxygen is in excess and the reaction temperature is not too high, prompt-NOx can be neglected, its contribution is less than 1ppm [Aguire, 1995]. Thermal-NOx is normally low for temperatures below 1000°C, but at high aeration rates, concentrations of molecular nitrogen and oxygen may play a significant role. The fuel-N is the most important as its depends directly on the nitrogen content of the waste. During devolatilization, partial nitrogen conversion to NH_3 is believed to be a good approximation for organic fuel (Jenssen and Johnsson, 1997; Johnsson and Dam-Johansen, 1995). Recent studies indicate that volatile HCN does not play a major role in the formation of NO and N_2O from combustion of char-N and NO is considered as the principal precursor of N_2O formation [Goel et al., 1996; De Soete, 1990; Johnson and Dam-Johansen, 1997]. Several authors have reported the influence of bed material and char on the reduction of NO and N_2O which explains the low gaseous emissions observed from fluidized beds [Orlanders and Strömberg, 1995; Tsuo et al., 1995; Johnsson et al., 1997; Li et al., 1998].

CO oxidation is the main carbon related reaction as other reactions involving NO and N_2O are less important for carbon due to the high ratio of C/N in the waste. The reaction of char-C+CO2→CO is thermodynamically negligible for temperatures lower than 1200°C [LaNauze,1985]. The system of reactions in Table 3.1 suggests that CO, char and bed material play a major part in the formation and destruction of NO and N_2O. The rate of formation of sulfur dioxide is proportional to the reaction of waste. At the same time SO_2 may be captured the presence of calcium oxide present in the bed as discussed elsewhere [Lin et al., 1995; Rajan and Wen, 1980]. It must be noted that most of the reactions in Table 3.1 involve superequilibrium concentrations of intermediate radicals such as OH, H, O and CH. The presence of halogen products in the waste may hamper some reactions and result in higher emission level [Anthony et al. 1993, 1995].

3.4.4 Volumetric Rate Equations of Species In the Combustor Zones

The mass balance Equations (1) require a kinetic rate model for each species. A general form of the reaction rate is summarized below with applicability to all reactor zones: the riser, the disengagement zone and the cyclone where the voidage is unity:

$$R_{NH3} = R_{Volatile} - 2R_3 - 2R_{14,CAT}$$

$$R_{NO} = R_3 + R_4 + R_5 - R_6 - R_{7,CAT} - R_8$$

$$R_{CO} = R_1 - R_2 + R_6 - R_{7,CAT} + R_{10} - R_{11,CAT}$$

$$R_{CO_2} = R_2 + R_{7,CAT} + R_{11,CAT}$$

$$R_{SO_2} = R_{12} - R_{13}$$

$$R_{H_2O} = 0.5F_H(R_{WASTE} + R_{CHAR}) + 1.5R_3 + 3R_{14,CAT}$$

$$R_{O_2} = 0.5(F_{O_2} R_{CHAR} - R_1 - R_2 - 2.5R_3 - R_4 - R_5 + R_{9,CAT} - 2R_{12} - R_{13}) - 1.5R_{14,CAT}$$

$$R_{N_2} = -0.5R_4 + 0.5(R_6 + R_{7,CAT}) + (R_{9,CAT} + R_{10} + R_{14,CAT})$$

$$R_{N_2O} = R_8 - R_{9,CAT} - R_{10} - R_{11,CAT}$$

The heat balance is presented elsewhere (Mukadi et al., 1998) and will not be repeated here.

3.5 Results and Analysis

3.5.1 Experimental analysis

Experimental work is performed to verify some model assumptions with respect to thermal treatment of foundry sand that have properties listed in Table 3.3. For this, combustion of natural gas is carried in four different ways: in an empty vessel, with clean sand in batch, and with feeding of clean sand and spent foundry sand. Empty vessel combustion of natural gas yielded less than 5 ppm for CO at temperatures above 900°C, more than 30 ppm of NOx at temperatures exceeding 1000°C even at lower aeration rates and SO_2 was at all time below 3 ppm. Figure 3.3 shows the data obtained as the bed is progressively heated by natural gas. These data also reveal that when temperature is beyond 900°C, NO emissions are high but CO is low. It is therefore suggested that the NOx are principally formed from thermal sources alone. The results on CO emission support the idea that temperature has the major effect on oxidation of CO. The effect of bed material (sand) on gaseous emission is shown. As expected, at low temperatures, CO emissions are high and NO are low because sand does not contain nitrogen. At temperatures higher than 1000°C, CO emissions drop sharply, NO emissions start to increase but stay lower as compared to the empty vessel results. Similar observation were reported by Bulewicz et al. (1997) who attributed that to prompt-NO formation. However, this may be explained by the fact that bed materials have some effect on NO reduction in fluidized beds as also supported by the work of Orlanders and Strömberg (1995) which attests that sand possesses some activity for reactions involving gaseous emissions; NO is believed to be a precursor of N_2O formation via the mechanism: Char-N→--CNO→ N_2O (De Soete, 1990). These two nitrogen oxides are reported to be reduced over bed materials such as ash+sand and char [Johnsson and Dam-Johansen, 1995]. From this discussion, it is concluded that the combustion of natural gas does not generate carbon monoxide provided the bed temperature is beyond 900°C. Hence, under these conditions, carbon monoxide from the burner does not interfere with the combustion of the waste. The kinetic of natural gas combustion is not included in the present model. Depending on operating temperature, thermal-NO may be produced, which is accounted for by reaction-4 in Table 3.1 and 3.2.

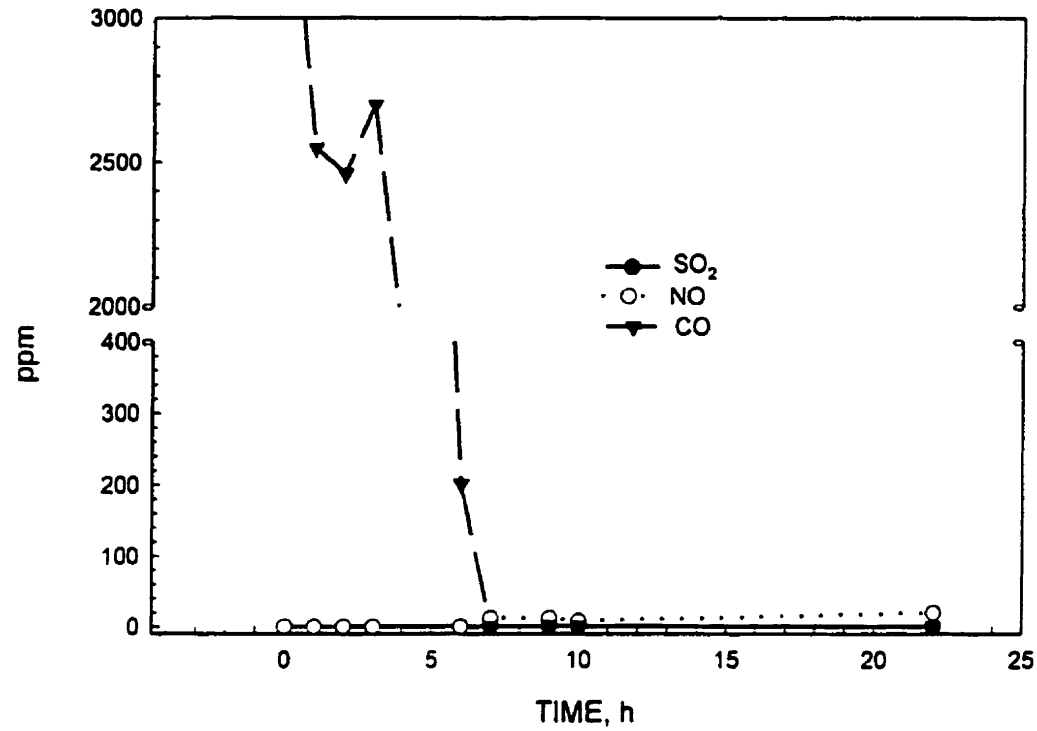

Figure 3.3: Experimental data for gaseous emissions during heat-up of sand bed in a 20 kW ICFB combustor; prior to actual thermal treatment of spent foundry sand. Solid circulation flux: 35 kg/m^{2}s; air flowrate: 36kg/h; aeration rate: 1.5. temperature from 700 to 1000°C.

Figures (3.4-a and 3.4-b) compare gaseous emissions obtained before and after the feed of spent foundry sand for two different resin concentration. CO emissions remain low as the temperature is beyond 900°C; however, NOx emissions are extremely high unlike in Figure (3.3) with similar operating temperatures. Yet considering the chemical composition of the foundry sand (see Table 3.3) and the waste feedrate, and converting completely waste-bound nitrogen to NO and sulfur to SO_2, one obtains values over 10000 ppm and 50 ppm respectively. The experimental data suggest that reducing reactions as underlined in Table 3.1 are responsible for low concentration of pollutants in the flue gas. According to these reactions carbon monoxide, bed material and char play a major role in the NO related reactions. CO and char favour the reduction of NO to N_2 and in case devolatilization is present, NH_3 simultaneously and catalytically transforms to NO and N_2; which in the overall produce low NO emission levels. Nevertheless, the higher resin contant sand (Figure 3.4-b) yields, as expected, higher NO than the lower resin content sand. The presence of NO and char-bound nitrogen reaction are believed to be the main route of N_2O formation the fluidized bed. The model computes N_2O according to the reaction mechanism proposed by Goel et al. (1994) and De Soete (1990).

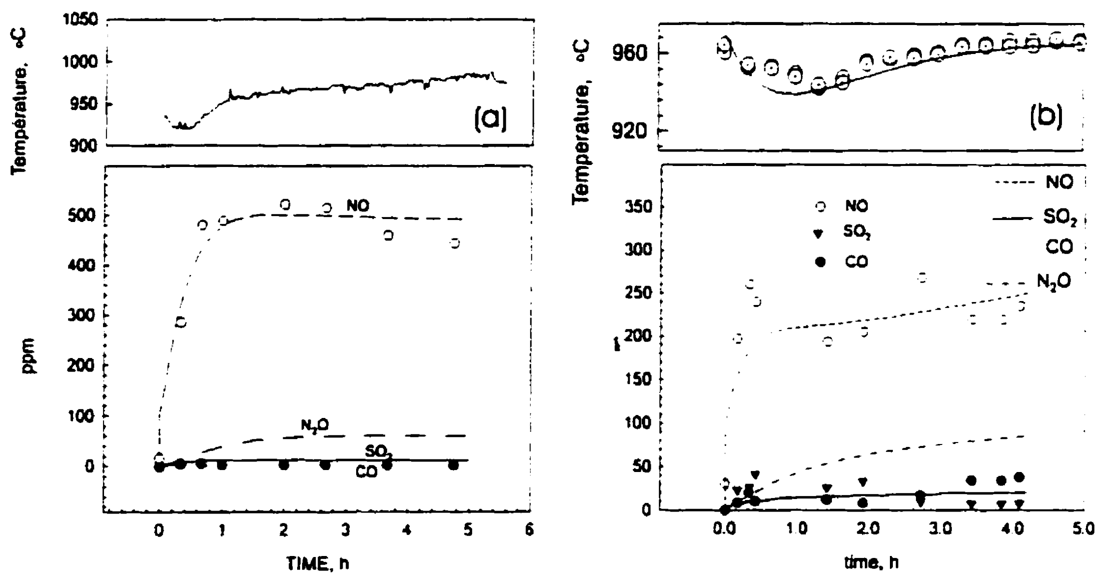

Figure 3.4: Comparison of experimental data and model prediction for treatment of spent foundry sand in a 20 kW ICFB combustor with temperature increasing from 920 to about 980°C. aeration: 1.45. at 12% O_2 (a) organic content 2%; solid circulation flux 38 kg/m^{2}s; air flowrate 36 kg/h; solid feedrate 35kg/h. (b) organic content 3.2%; solid circulation flux 35 kg/m^{2}s; air flowrate 36 kg/h; solid feedrate 25kg/h.

3.5.2 Model Validation

The model uses the reaction kinetics of Table 3.1 to estimate concentration of CO, NO, SO_2, O_2, and combustion efficiency for a range of operating conditions. Experimental data used for model validation were obtain during spent foundry sand combustion in a 20 kW ICFB unit. Thermogravimetric analysis have shown that volatiles are negligible in the spent foundry sand. In addition, the model is used to analyze the effect of some operating parameters. The NO and N_2O related reactions are strongly affected by the char content and bed material. In the proposed model, volumetric char content is proportional to organic concentration within the bed. The reactivity of char and bed material depends on its chemical and physical properties, [Li et al., 1998; Johnsson et al., 1995; De Soete, 1990]. This variation may even attain three orders of magnitude [Liang and Kristiina, 1998]. In the present model the difference in char and bed material reactivity is taken into account by using an activity factor addition to the literature data. In this modeling, the reaction rate coefficients are multiplied by a factor of 65 for heterogeneous reactions involving char. The model accounts for sulfur capture due to the small amount of calcium oxide present in the bed material.

Amongst industrial wastes potentially capable of being treated in an ICFB, spent foundry sand can be regarded as a model waste. Its chemical and physical properties are relatively constant when compared to other wastes. In spite of this high degree of uniformity, variation must be expected from the experimental results, which render model validation difficult. However, the purpose of the model is not to be highly accurate but rather to be able to describe essential phenomena, yielding comprehensive predictions for concentrations of pollutants in the flue gas. Accordingly, Figure (3.4-a, 3.4-b) show reasonable agreements between experimental and calculated results for treatment of foundry sand under unsteady state conditions, with operating temperature are between 900 and 1000°C. The model predictions follow the general trend displayed by the experimental data. The model shows that for temperatures above 900°C, CO emissions are low; the amount of nitrogen converted to NOx and N_2O is much lower than the actual amount of waste-bound nitrogen, which means that reduction reactions take place. Oxidation of CO is much faster and this result in very low emission levels. The predicted N_2O emission level is about 50 ppm in the range of values produced fluidized bed combustion (20-250 ppm) [Li et al., 1998]. Besides, N_2O is more readily destroyed than NOx through thermal decomposition and over bed material, this results in lower emission levels.

Table 3.3: Proximate and ultimate analysis of spent Foundry sand resin

Proximate, wt %	Ultimate, wt %
Foundry Sand Resin:	C : 69.33
Resin concentration, 2.06~3.2	H : 4.711
Moisture :0.19	O : 21.47
Volatile :<<0.01	N : 3.28
Ash : 97.75 (silica sand)	S : 1.24

Mean size, μm	180-190
Density, kg/m^3	2600
HHV, MJ/kg resin	26.56
Silica sand composition%(wt):	
SiO_2	96.4
TiO_2	0.00
Al_2O_3	0.15
CaO	0.10
MgO	0.03
Na_2O	0.015
K_2O	0.029
Fe_2O_3	0.207

3.5.3 Sensitivity analysis

Sensitivity analysis is performed using the model to see the effects of some operating parameters on the concentration of CO, NO, N_2O, SO_2, O_2 in the flue gas. The set of parameters used for this analysis is listed in Table 3.4. For analysis purpose, simulation is performed for treatment of foundry sand. One must not that all parameters are not independent as they relate to each other through heat and mass balances and kinetics in the ICFB.

Table 3.4: Standards scale-up simulation conditions

Riser: diameter 30 cm; height 3.1 m
External cylinder: diameter 720 mm; height 5 m
Solid inventory: 1355 kg
Air flowrate: 540 kg/h
Aeration: 1~2.6
Solid circulation flux: 35~100 kg/m^2s
Waste feedrate: 600~2000 kg/h
Pressure: 101.3 kPa

Effect of position. Simulations of axial profiles illustrated in Figure (3.5-a,b) show the fate of gaseous emissions along the reactor height. The simulations are performed with riser temperature increasing in the range of 900 to 1000°C. It is seen that CO and NOx increase quickly at the riser entrance and exhibit a maximum. As z increases CO disappears rapidly due to homogeneous reaction. This in turn slows the NOx catalytic reduction rate by CO. As the flue gas leaves the riser, with gas-solid separation, no more heterogeneous reactions occurs; the NOx remain almost unchanged because its principal reduction reaction no longer takes place; also temperature and oxygen concentration are not high enough to generate thermal NOx. The N_2O on the other hand, are primarily formed within the bed and afterward reduced by homogeneous thermal decomposition outside the riser. One may state that longer residence time within the disengagement zone and cyclone favors lower emission levels of N_2O and CO, whereas NOx is best destroyed heterogeneously inside the riser in presence of CO and char. SO_2 emission levels also depend on the heterogeneous reactions within the riser, in the present simulation only calcium oxide present in sand is assumed to contribute to sulfur capture as no additional calcium oxide is used. However, it is unlikely that the CaO of sand captures sulphur. The very low SO_2 observed may include significant experimental error as the treated foundry sand contains very little sulphur.

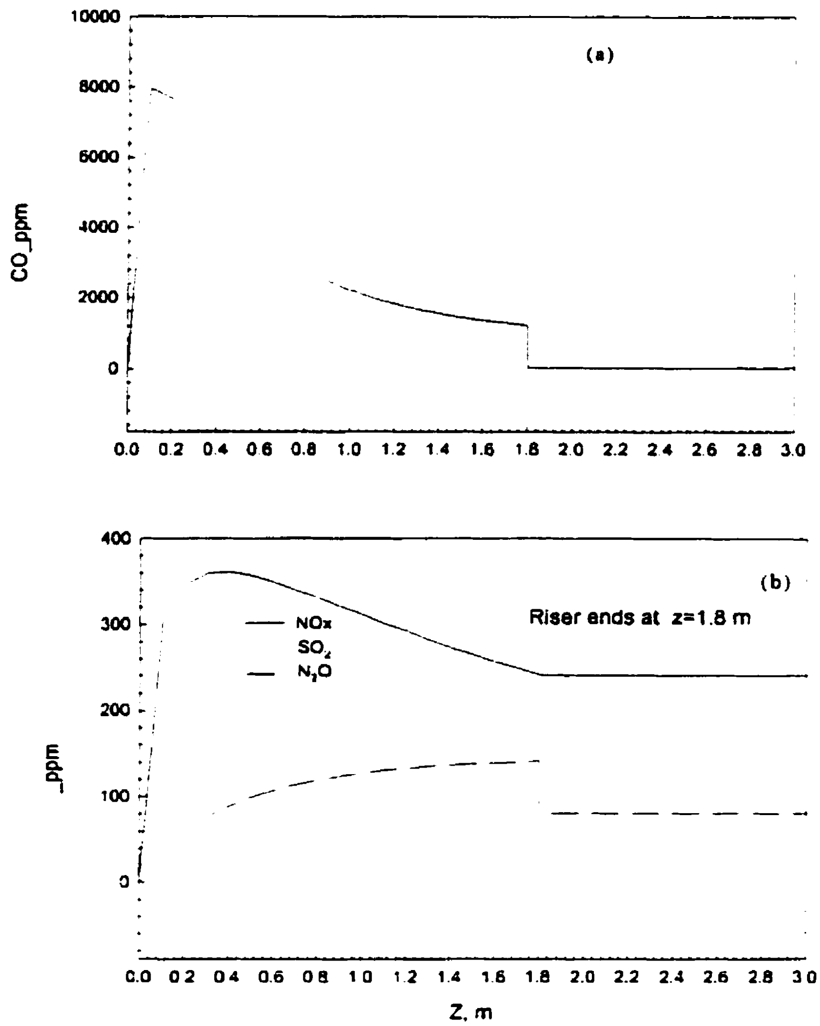

Figure 3.5: Prediction axial profile for the concentration of gaseous emissions at organic content 2%; solid circulation flux 38 kg/m^{2}s; air flowrate 36 kg/h; solid feedrate 35kg/h with temperature increasing from 920 to about 980°C, aeration: 1.45.

Effects of solid circulation flux. Figure (3.6) presents CO, N_2O and NO emissions computed at two different solid circulation flux as a function of solid feedrate. Increasing solid circulation decreases the voidage resulting in higher gas-solid contact area, which facilitates heterogenous reactions. Hence, NOx is readily reduced by char and bed material while increasing the formation rate of N_2O from char combustion; at the same time, heterogeneous destruction rate of N_2O increases which overall, results in lower emission levels. In earlier work by Mukadi et al. (1998), it is observed that high solid circulation fluxes produce high solid waste conversion by increasing the number of passes through the riser. As solid circulation is increased, voidage decreases resulting in more contact area, char content and bed material in the riser, which is propitious for catalytic and reduction reactions responsible for lower emission levels. However, CO is less affected by solid circulation as its main reduction reaction proceeds in the gas phase. Nevertheless, the sensitivity of CO concentration to solid circulation depends strongly on the ratio of C/N in the waste. Most waste of organic origin have very high ratio of C/N, hence nitrogen related reactions are less important for CO elimination.

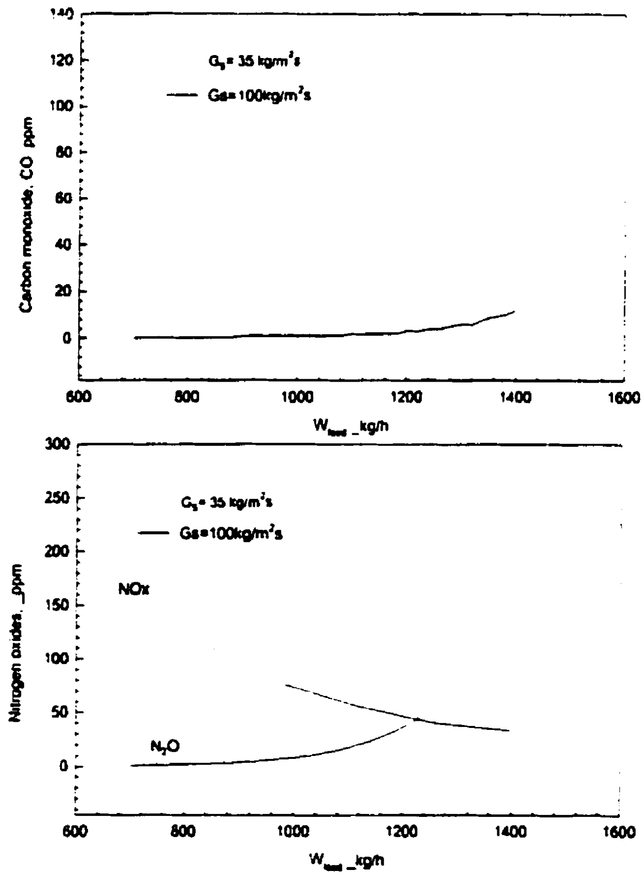

Figure 3.6: Simulation and prediction under scale-up conditions for gas emissions at different waste feedrates and circulation flux. Riser diameter : 30 cm, riser height : 3.1 m, external cylinder diameter : 72 cm, external cylinder height : 5 m. air flowrate : 540 kg/h, aeration rate : 1.7

Effects of waste feedrate. Figure (3.6) also shows predictions under different waste feedrates for a fixed aeration rate of 1.7. At low feedrates, temperature and oxygen concentration are high due to little organic concentration and less cooling effect. This leads to lower char content and lower CO concentration in the combustor. At this condition of high temperature, more thermal-NOx is formed. Increasing the feedrate decreases the temperature in this case, increases the organic and the char content within the bed, produces faster heterogeneous reaction rates. Less N_2O is destroyed as the temperature decreases and, more NOx is transformed into N_2O. When NOx reach the minimum, destruction and formation balances; this may be used for optimization purposes since beyond this point, temperature continues to decrease. When feedrate is very high, temperature becomes too low to sustain any reaction.

Effects of aeration rate. The aeration rate affects temperatures and oxygen concentration within the combustor. Very low aeration rates will result in lower oxygen concentration and result in inadequate combustion process. Extremely high aeration rates in the other hand may result in very low temperature and poor combustion also. Figure (3.7) shows simulations starting below the stoiechiometric aeration rate with respect to overall stoichiometry including resin. Literature data suggest that increasing aeration rate increases emissions of nitrogen oxides [Goel et al. 1995]. The present model predicts also similar trends at oxidation conditions as NO and N_2O have a nearly linear increase despite the decreasing temperature. This produced by the diminution of char content as combustion proceeds in presence of higher oxygen concentration levels. The system of reactions presented in this paper show that NOx and N_2O are destroyed catalytically and heterogeneously over char. Consequently, the diminution of char content in the bed decreases the NO and N_2O destruction rate and produces higher emission levels.

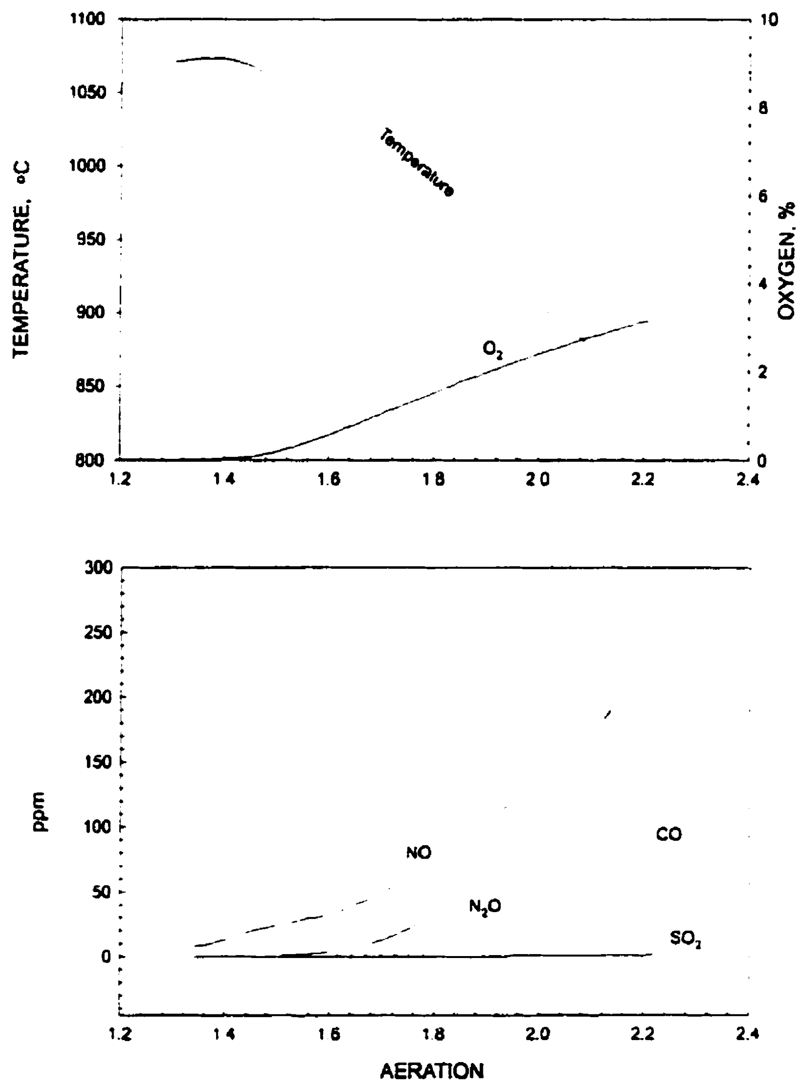

Figure 3.7: Influence of aeration rate on the emission levels of carbon monoxide, nitric oxide, oxygen concentrations, and temperature. Riser diameter : 30 cm, riser height : 3.1 m, external cylinder diameter : 72 cm, external cylinder height : 5 m. air flowrate : 540 kg/h, aeration rate : 1.7

3.6 Conclusion

A simplified model is proposed to predict the pollutant gas concentrations of an ICFB during thermal treatment of industrial wastes. The trends of model predictions are in qualitative agreement with data from the pilot scale ICFB combustor. The model can be used as a practical tool for a quick estimation of flue gas concentrations of CO, NO, N_2O, SO_2, O_2, and CO_2, and for preliminary estimation of operating parameters.

The gaseous emission levels are dependent on waste feedrate, aeration rate, solid circulation and waste properties. The CO oxidation is the main route of CO elimination. The kinetic model indicates that only a number of homogeneous and heterogeneous reactions are important during the formation and destruction of gaseous emissions in the ICFB combustor. The presence of bed material such as char and ash+sand plays a significant part in the reduction of nitrogen oxides (NOx and N_2O). The model also shows that emission levels observed in the ICFB are a balance of formation and destruction reactions. It can be used for quantitative and qualitative modeling and design purpose.

3.7 Acknowledgement

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Chapitre 4 PARAMETER ANALYSIS AND SCALE-UP CONSIDERATIONS FOR THERMAL TREATMENT OF INDUSTRIAL WASTE IN AN INTERNALLY CIRCULATING FLUIDIZED BED REACTOR

Reference: Lukanda Mukadi, Christophe Guy*, Robert Legros (1998): "Parameter Analysis And Scale-Up Considerations For Thermal Treatment of Industrial Waste In An Internally Circulating Fluidized Bed Reactor », submitted *to Chemical Engineering Science*.

Keywords: Circulating Fluidized Bed, Combustion, Solid Waste, Simulation, Scale-up, Modeling

* Author for correspondence, E-mail address: Christophe.Guy@mail.polymtl.ca

4.1 Contexte[a]

Les chapitres deux et trois proposent un modèle pour le traitement thermique de déchet dans le LFCI. Lorsque la formation des polluants gazeux durant le traitement thermique n'est pas d'une grande importance, le chapitre deux tout seul est suffisant pour décrire le comportement de l'unité. Cependant, le chapitre trois complète le modèle du LFCI en le dotant des capacités d'exhiber les comportements additionnels relatifs aux contaminants gazeux.

La performance du LFCI est fortement fonction des conditions opératoires et des paramètres géométriques. Ainsi, pour avoir une performance optimale, un design efficace et une stratégie adéquate sont requis.

Le modèle présenté au chapitre deux et trois est utilisé dans ce nouveau chapitre pour analyser les effets de différents paramètres sur le comportement du LFCI afin d'identifier et de déterminer certains critères et stratégies pouvant servir au design et à l'opération efficace de l'unité.

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[a] ne pas inclut dans l'article

4.2 Abstract

A model was previously developed to predict the combustor performance of an Internally Circulating Fluidized Bed Reactor (ICFB). The model was validated with experimental data obtained during the thermal treatment of different industrial wastes in a 20 kW ICFB pilot unit. The model gave good agreement with experiments without the use of adjustable parameters. Simulation was made to analyze the behavior of reactor depending on different design and operating variables as well as waste properties. The variables most affecting the ICFB performance were solids circulation flux, gas-solid feed rates, riser diameter, working bed temperatures and waste properties. The ratio, G_sA_r/W_{feed}, is a critical parameter for design and operation.

4.3 Introduction

Circulating fluidized beds have made large impact in catalytic processes, in coal combustion technology and in biomass or waste thermal treatments. They have the advantage to achieve greater efficiencies in short gas-solid contact times, and they offer the possibility to control separately the retention times of gas and solids. The success of circulating fluidized beds combustion lies in its low gaseous emissions and the ability to burn a variety of fuels including low grade combustibles. The internally circulating fluidized bed (ICFB) tends to be shorter and compact. One of its major current application is thermal treatment of industrial solid wastes [Lovett et al., 1997; Mukadi et al., 1997].

On-site treatment of wastes with mobile units is increasingly adopted as a way of reducing costs while meeting environmental regulations [McGowan and Ross, 1991; Baukal et al., 1994]. The internally circulating fluidized bed, due to its compactness, is appropriate as mobile unit. However, proper engineering design and operating strategies are required for such a transportable or mobile unit as it will be frequently operated under unsteady state conditions and with different types of wastes. It is therefore important to provide transient models for predicting start-up and shut-down periods, calculating response to upset conditions and developing control strategies. Several attempts made to model the CFB deal exclusively with what happens at steady state conditions inside tall risers [Grace and Lim 1997] for which the lower acceleration zone is negligible. A complete model for ICFB treatment of industrial waste should describe the internal phenomena within the riser, the disengagement zone and the annular bed, and should simulate both steady state and transient conditions.

Weiss and Fett (1986), Weiss et al. (1987, 1988), Mori et al. (1991), Zhang et al. (1991), Hyppanen et al. (1993) and Muir et al. (1997) have developed transient models for coal combustion processes. These models subdivide the riser (and the external loop) into several cells in series and the lower dense zone is treated as a bubbling bed or ignored. Each cell is assumed to be perfectly mixed with some allowance of backmixing between cells. Though these models meet current needs, they are oversimplified and they can not be directly applied to simulation of the ICFB for treatment of industrial wastes. This paper presents an ICFB simulation adopting the model developed by Mukadi et al.

(1998) which accounts for dynamic changes of hydrodynamic properties, gas-solids temperatures and concentrations. The influence of key model parameters is studied, and some scale-up and operating strategies are proposed.

The experimental unit in Figure 4.1 is made of two concentric cylinders—the riser and the external cylinder which contains the annular bed. A screw regulates feedrate of fresh solids which are transported pneumatically into the riser. Fresh solids and solids from the annular bed mix at the riser base, and are carried up by the flowing gas. At the riser top, the presence of an impact device and the disengagement zone provide gas-solids separation. The gas and a small amount of particles fine flow out to the cyclone, while the separated solids move downwards through the annular bed to be re-injected at base of the riser.

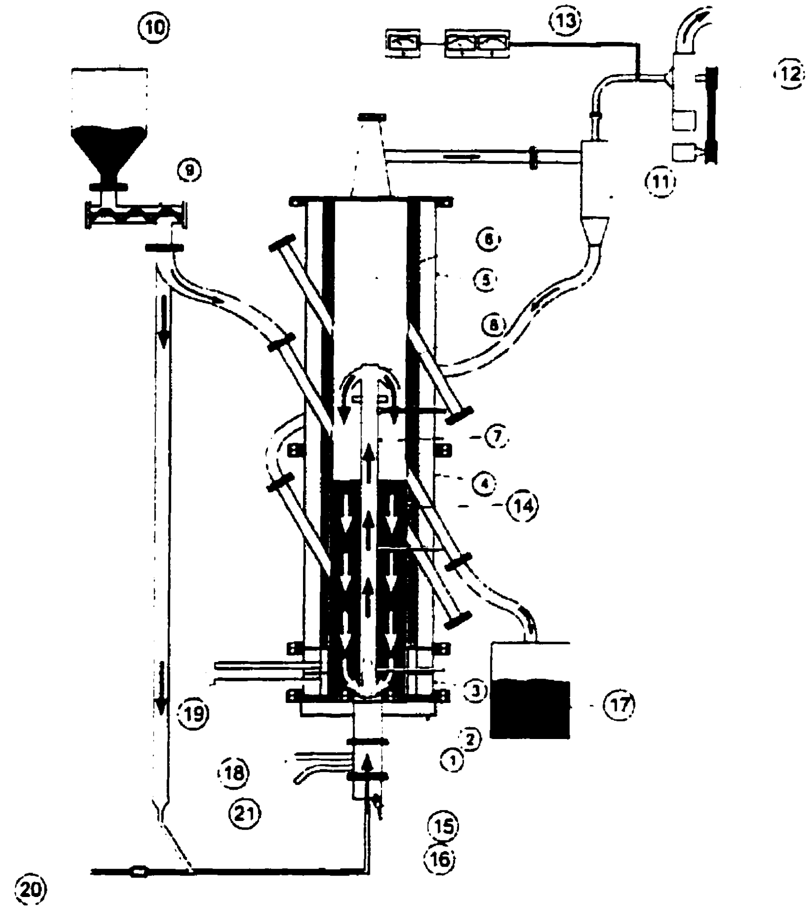

Figure 4.1: Schematics of the Internally Circulating Fluidized Bed pilot unit:

1-Plenum, 2-Support plate, 3- Secondary air distributor, 4-Lower chamber, 5-Upper chamber, 6-Disengagement zone, 7-Riser, 8-Impact device, 9-Screw-conveyor, 10-Hopper, 11-Cyclone, 12-Ventilator, 13-Flue gas analyzer, 14-Annulus region, 15-Burner, 16-Spark plug, 17-Treated solids, 18-Primary air, 19-Secondary air, 20-Air for pneumatic transport, 21-Natural gas. Riser --length: 1.5m/diameter: i.d.780mm, o.d.890mm. External cylinder-- length: 2.5m/ diameter: i.d.355mm, o.d.645mm. Burner length: 300mm

4.4 Simulation and parameters analysis

The benefit of simulation is in enabling off-line investigation between model parameters and process performance. This is advantageous for elaborating operational methods and design criteria. ICFB parameters can be classified in three categories: — operating parameters, which include gas and solid feed rates, aeration rates, circulation flux, bed temperature, and solid inventory — design parameters, referring to bed geometry: height and diameter of both the riser and the outer cylinder — waste characteristics consisting of chemical and physical properties, particle size distribution, organic, volatile and moisture contents. To study the effect of waste type, the model developed by Mukadi et al. (1998) was reformulated to account for evaporation rates and particle size distribution was characterized by the use of a mean particle diameter. The mass balance in the riser can be written as:

$$\frac{\partial Y_{pr}}{\partial t} = V_p \frac{\partial Y_{pr}}{\partial z} - \sum_{i=1}^{n} R_{s,i} \quad ; \quad V_p = \frac{G_s}{\rho_p (1-\varepsilon)} \left(1 + \frac{W_{feed}}{G_s A_r} \right) \tag{1}$$

The heat balance can also bemodified to take into account the heat consumed by evaporation. Some important additional parameters are noticed—the ratio G_sA_r/W_{feed}, and the mass flux W_{air}/A_r which represents the superficial gas velocity. $R_{s,i}$ includes reaction rates and evaporation rates. The number of passes through the riser can be estimated as:

$$N_{pass} = \frac{\text{Overall residence time}}{\text{one pass residence time}_{(\text{Riser+annular bed})}} \cong \frac{G_s A_r}{W_{feed}} \tag{2}$$

The value of N_{pass} is important for scale up, as it will be used to determine the geometrical and operational parameters of the ICFB unit.

4.4.1 Effect of waste type.

Different types of industrial wastes have been thermally treated in the ICFB to demonstrate its flexibility [Boisselle et al., 1998; Mukadi et al., 1997]. In this study, consideration is given to thermal treatment of such wastes as spent foundry sand, deinking sludge and contaminated soil. Spent foundry sand is a representative of a dry waste with uniform particle size and a low heating value. Pre-dried deinking sludge is typical of waste with a substantial organic content hence higher heating value. Contaminated soil relates to non-uniform size solid wastes with high moisture and very low organic contents.

4.4.1.1 Contaminated soil

Contaminated soils used here have a particle size distribution of 0.3 to 5mm and about 10% of moisture content. They are representative of an actual PCB contaminated soil with less than 1% of organic material and a very low heating value. An average of 1mm was taken for the simulation. With treatment conditions, high temperature differences exist between the gas and solid phases and heating of particles is very fast. Also, solid circulation create a high convective heat flow between the riser and the annular bed, appropriate for high heat transfer and rapid evaporation. It is assumed that the drying rate in the riser is constant and equal to the gas-solid mass transfer rate, and complete drying is achieved in the annular bed. The model assumes that the drying particles are heated in the riser and enter the annular bed to reach complete drying due to higher particle residence time. Figure (4.2a) shows predicted and experimental data for the mean particle size of 1mm, and temperatures are average values in each zone. The pics are the result of changing the feedrates of gas and solid. The results show reasonable agreement with the experiments. Despite solids circulation, temperature difference between the riser and the annular bed is larger which may be attributed to lower heat transfer coefficients resulting from larger particle size. The simulation input parameters are natural gas and solid feedrates, hence the calculated temperature fluctuations are due to variations in these two parameters.

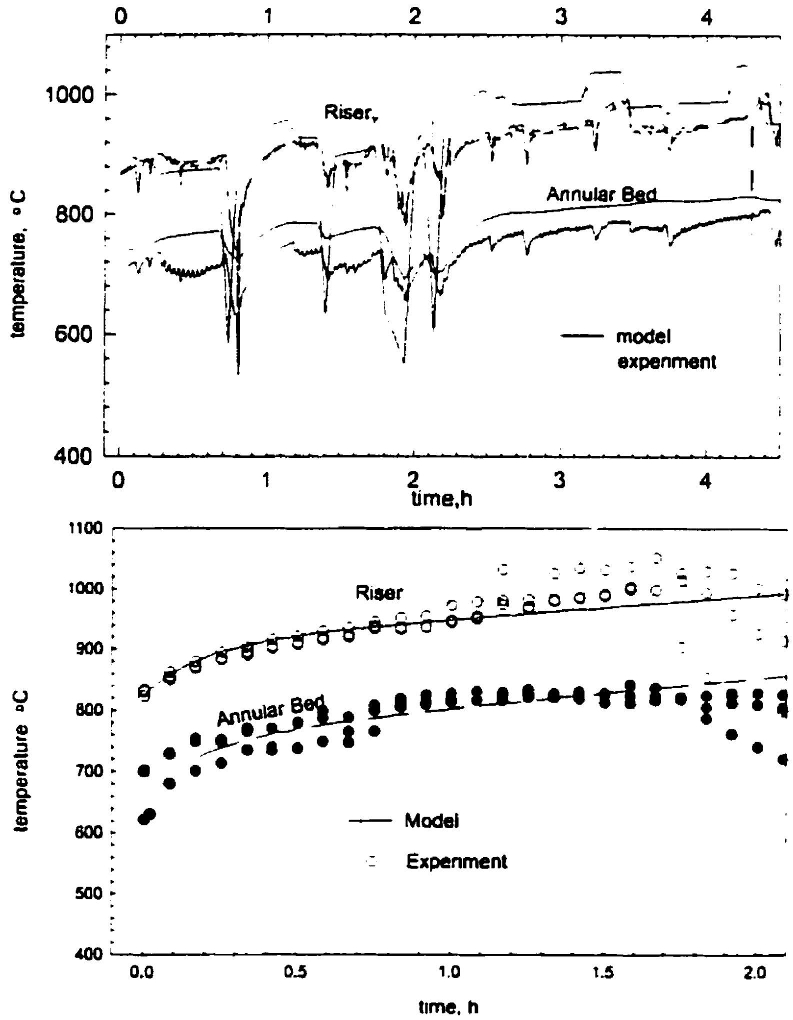

Figure 4.2: (a) Comparison between experiment and computed temperature profile for treatment of contaminated soil, $M_{s,tot}$= 100 kg, Gs= 30 kg/m^{2}s, total mass fed: 228 kg. (b) Temperature profile during Deinking Sludge treatment, Air flowrate≈ 37.4 kg/h; Aeration= 1.87, Initial organic content= 47%; X_{exit}>94%.

4.4.1.2 Pre-dried deinking sludge

Treatment of deinking sludge is of major importance to the pulp and paper industry. Sludge from a typical paper recycling industry was dried and its composition is listed in table (4.1). The feedrate was calculated to keep the bed temperature lower than

1100~1200°C. Simulation was performed adopting the kinetics from the work of Chang et al. (1996). The model computes bed temperatures, organic destruction and oxygen concentrations. In Figure (4.2b) the computed temperatures in the riser and the annular bed indicate the behavior of such waste in the ICFB. The pre-dried sludge contains about 50% of organic material and a large heating value. The treatment of such wastes require less auxiliary fuel to sustain combustion. As before, the simulation input parameters are the natural gas and sludge feedrates.

Table 4.1: Composition of pre-dried sludge.

Proximate, wt %	Ultimate, wt %
Fixed carbon : 2.73	C : 25.62
Moisture : 2.15	H : 2.62
Volatile : 47.90	O : 25.12
Kaolin ash : 47.22	N : 0.14
Density: 308 kg/m^3	S : 0.04
Heating value: 14.0 MJ/kg (ash free)	

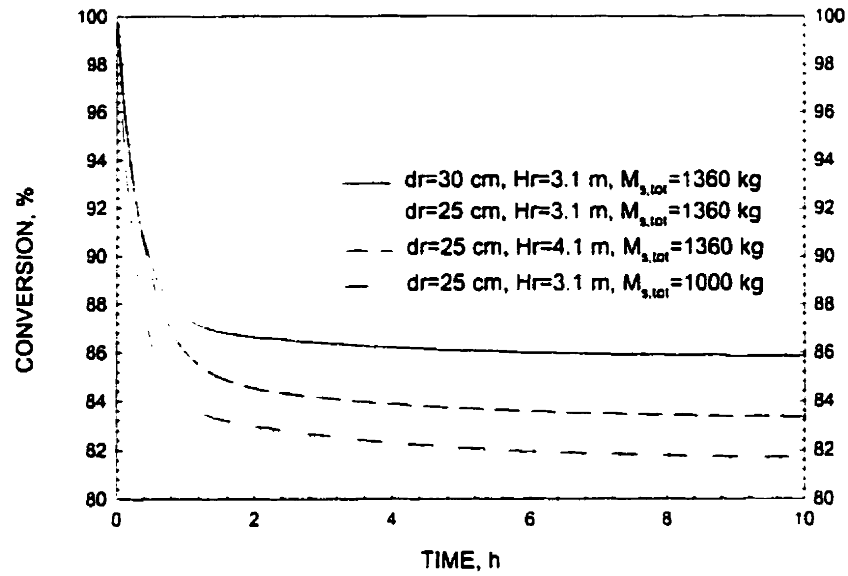

Figure 4.3: Transient prediction of conversion using different bed sizes to analyze the effect of riser diameter, riser height, external cylinder diameter and total solid mass, W_{feed}=1000 kg/h, W_{air}= 540 kg/h, n=1.7.

4.4.2 Effect of Bed Geometry

Commercial dimension of CFB combustors can be up to 3~10 m in diameter and 15~50 m in heights (Zhu and Bi, 1995) as high energy output is required. However, the ICFB unit being developed for waste treatment, is suited to be small and compact. From former work by Mukadi et al. (1998), a rough estimate suggests that an equivalent riser diameter less than 0.40 m is sufficient to treat over one ton of waste per hour. The riser diameter is linked to flow rates (G_sA_r and W_{feed}) and strongly affects the number of passes through the riser (N_{pass}). Its height is an important design parameter which influences the mean particle residence time and the overall treatment performance. The external cylinder determine the volume of the annular bed, which provides particles for circulation and acts as thermal inertia. Simulation analysis were performed with the set of parameters presented in Table (4.2). Figure (4.3) indicates that the riser diameter has a strong effect on the conversion. Initially, there is no organic material in the bed, hence conversion starts at 100%. Increasing the riser diameter increases the mass flowrate (G_sA_r), which greatly reduces the particle residence time in the annular bed in favour of the riser. Therefore, this increases the number of passes across the riser (N_{pass}) which overall, results in higher conversion. Figure (4.3) reveals that a taller riser only produces a small conversion increase despite the higher riser residence time per pass. Only very high increase in riser height would have a similar effect as the riser diameter on conversion. This is because the total residence time of particles in the riser is a very strong function of the riser diameter as compared to riser height. The external cylinder diameter has little effect on the overall conversion. This implies that a smallest possible annular bed volume should be kept for the sake of particle circulation and stocking of heat. Small annular bed volume would be most appropriate for rapid bed heatup and rapidly reaching steady state operation, which is important for a unit that needs frequent shut-downs and start-ups or load changes. The disengaging zone and the impact device at the riser top play a major role for gas-solid separation in the ICFB. A proper design minimizes the need of an external cyclone which is not addressed in the present simulation analysis.

Table 4.2: Set of parameters used in simulation analysis

Geometry	Operation
Geometry: height & diameter	W_{air}: 540; 670 kg/h
Riser: H=3.1, 4.1; d=0.25, 0.30	n: 1~3.3
outer cylinder: H=5; d=0.72, 0.60	Gs: 35, 100 kg/m2s
Construction material thickness:	W_{feed}: 200~2000 kg/h
refractory: 6.35 cm	$(m_{s,tot})_{max}$: 1360; 940 kg
mineral wool: 7.62 cm	
steel: 0.5 cm	

4.4.3 Effect of operating conditions

Thermal treatment of any type of waste depends on proper operating conditions which affect conversion, bed temperature, residence time and gaseous emissions. Effects of such operating variables are analyzed with respect to the thermal treatment of foundry sand. Foundry sand is used to make mold in metal casting industry. It is a mixture of organic binder and sand. It can be reused several times before loosing its molding capacity and become spent foundry sand. Today, its reclamation starts to make economic sense due to increased landfill costs and severe environmental regulations.

4.4.3.1 Effect of solids circulation

Thermal treatment of solids is generally not achieved after a single pass into the riser where particle residence time is in the order of 1~3 s. Highly organic wastes or humid waste which require longer processing time can be thermally treated in the ICFB by virtue of solid circulation and the presence of the annular bed which assures circulation.

Moreover, solids circulation of high heat capacity solids creates high convective heat flows responsible for small temperature gradients in the riser and the annular bed. Similarly to the riser diameter, the solid circulation, Gs, affects residence time in the ICFB thus affecting N_{pass} and the conversion. In Figure (4.4a), it is shown that conversion increases with Gs. This results from the increase in N_{pass}, similar to the effect of the riser diameter.

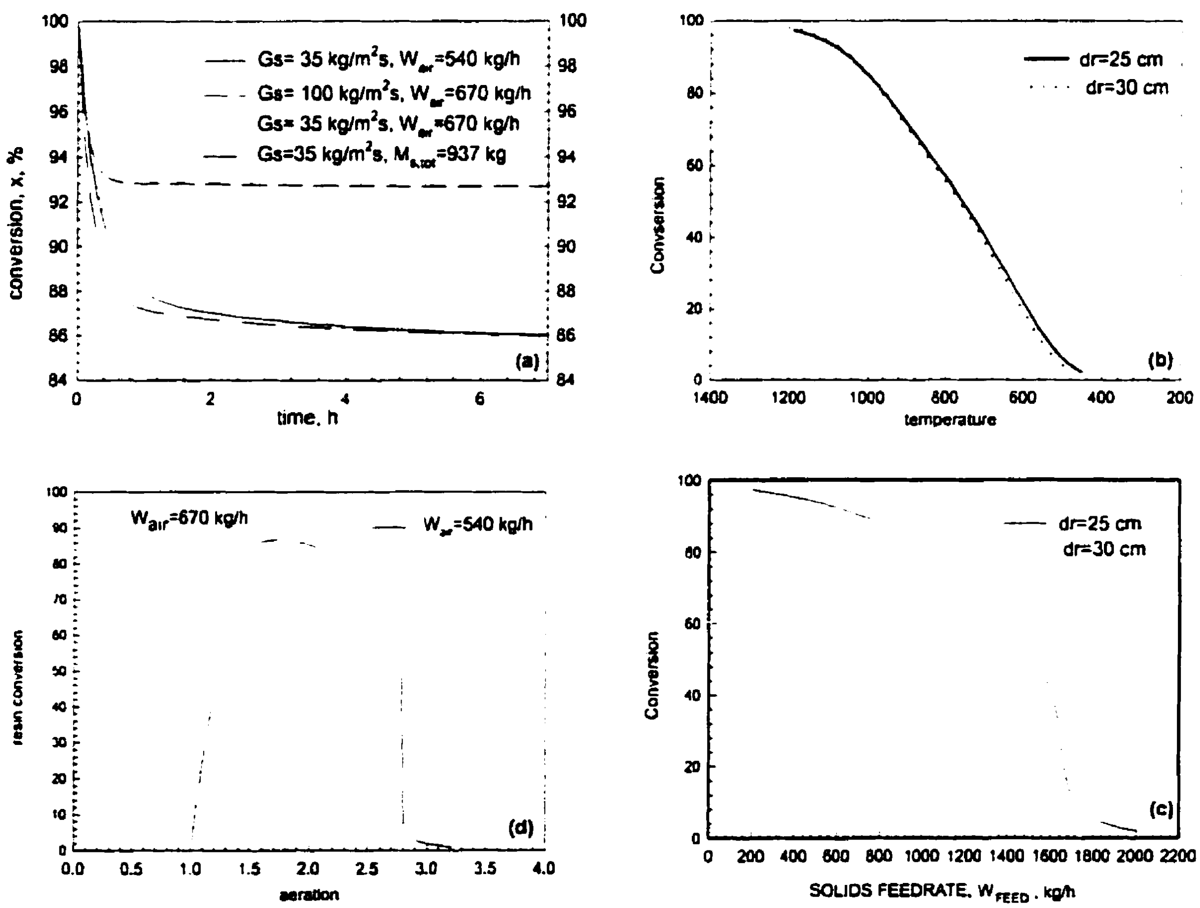

Figure 4.4: $M_{s,tot}$=1360 kg, d_{riser}=0.30 m, H_{riser}=3.1 m, $d_{cylinder}$= 0.72 m. (a) Transient prediction to show the effect of solid circulation flux, Gs, W_{feed}= 1000 kg/h, W_{air}= 540 kg/h, n=1.7; (b, c) Effect of solid feedrates and riser diameter on the temperature and the conversion, W_{air}=540 kg/h, n=1.70; (d) Effect of air flow rate and aeration rate on the conversion.

4.4.3.2 Effect of solid waste feed rates

The solids feedrates of interest for spent foundry sand treatment are in the range of 800~2000 kg/h. For a fixed bed diameter, there exists a maximum feedrate that guaranties good performance. The solids feedrate controls the overall residence time, $m_{s,tot}/W_{feed}$, which shows the importance of annular bed inventory. In Figure (4.4b,c), it is seen that conversion decreases with an increase of W_{feed} due to the decrease in overall residence time. In contrast, decreasing $m_{s,tot}$ diminishes the overall residence of particles without significantly lowering the conversion, as in Figure (4.3). This due to the fact that changing $m_{s,tot}$ does not affect what occurs in the main reaction—the riser. Simulations presented in Figure (4.4b,c) displays a S shape and an extinction feedrate or "blowout velocity" as solids feedrate is increased. The operation should be set below the blowout velocity in accordance with the desired conversion, emission standards and optimal bed temperature. The figures reveal that wider operating conditions can be used with larger diameters. Figure (4.4b,c) also illustrates that with two different diameters and the temperature remaining constant the conversion undergoes changes that can only be attributed to an increase of $N_{pass}(=G_sA_r/W_{feed})$.

4.4.3.3 Effect of aeration and air flow rates

Adequate air flowrates and aeration are needed to maintain high superficial gas velocities in the riser (4~15 m/s) and to provide oxygen for combustion. The superficial velocity depends on bed temperature and gas flow rate. It affects the solids residence time in the riser but it can not be studied as an independent variable except through gas flow rate. The bed temperature and the oxygen concentration are intimately linked to both aeration rate and air flowrates. Stoichiometric air feedrates produces high temperatures (>1200^{0}C) and very low oxygen concentrations, resulting in incomplete combustion of waste, and possibly in fusion or agglomeration of bed material. Higher aeration would lower bed temperature and slow the reaction. Figure (4.4d) demonstrates the existence of an optimal region for which conversions are optimal. At very high aeration, extinction conditions are reached, which correspond to a sharp fall in the conversion and temperatures. It can also be observed that for higher flow rates and constant aeration rate, superficial gas velocity is higher and particle residence time in the riser per pass is shorter. However, oxygen concentration and fuel input remain high enough to overcome this reduction in residence time, which results in higher conversions. Practically, very high aeration will have adverse effect on the formation of thermal NO_x as a consequence of enriched oxygen concentration. Hence, it is necessary to find optimum operating air flow rates and aeration rates accounting for organic destruction, gaseous emissions and bed temperatures.

4.5 Design Procedure

As observed from the experimental and simulation results, the performance of an ICFB combustor depends on several variables. A design procedure determines the reactor size and specifies the operational limits for the unit are presented in Table (4.3). Step 1 serves to determine the riser height and diameter from knowledge of N_{pass}, Gs and intended maximal solid flowrate, W_{feed}; for compactness, the riser should be as short possible. Step 2 is used to compute the size of the external cylinder which is to contain the annular bed and the disengagement zone. For convenience, the annular bed height should bed slightly shorter than the riser height. Annular bed mass has been shown to have a very small effect on the performance, and it is advisable to use values which result in short overall residence time and rapid heat-ups. From the riser diameter, annular bed height, solid feed rate and residence time of about 0.5~1 h, the external cylinder diameter is calculated. Because an impact device is used for solids separation, the disengagement zone on the riser top can be short, in the range of 1~2 m. However, the diameter in this upper zone a bit larger to keep superficial gas flowrate below that of the minimum fluidization. Step 3 estimates the operating air flowrate by selecting the riser gas superficial velocity and temperature. Finally step 4, which computes conversion and temperature as a function of aeration rate, is used to find the optimal operating conditions while maintaining temperature between 850~1200°C. These optimal conditions depend on different factors as underlined above, including organic and moisture contents.

Table 4.3: Proposed design steps and strategies for an ICFB unit

Step 1: riser size	**Step 2**: outer cylinder
Given: W_{feed}, Gs, N_{pass} *find* d_{riser} *from*: $\frac{G_s A_r}{W_{feed}} \approx 10 \sim 15$ riser *height*: $H_{riser} \approx (9 \sim 15) d_{riser}$	*Annular bed height*: $H_a \approx H_{riser} - .15\,m$ *find diameter from*: $\frac{M_{s,tot}}{W_{feed}} = \frac{\pi}{4}\frac{A_a \rho(1-\varepsilon)H_a}{W_{feed}}$ $\cong 0.5 \sim 1\,h$: with $A_a = \frac{\pi}{4}(d_{annular} - d_{riser})$ *cylinder height:* $H_{cyl} = H_{riser} + (1 \sim 1.5\ m)$
Step 3: air flowrate From superficial gas velocity of 8~12 m/s, temperature 1200°C: Compute: W_{air}	**Step 4**: optimal points *Calculate*: $X_{conversion} \sim n$ $T_{temperature} \sim n$ *Constraint*: $850 < T < 1200°C$

4.6 Conclusion

A validated mathematical model was used to predict the performance for an ICFB thermal treatment of industrial wastes. The model gives reasonable predictions under practical operating and design conditions, without the use of adjustable parameters.

The simulation analysis produced information on critical variables for design and operation of an ICFB reactor for treatment of industrial waste. In this sense, the strong effect of waste type, solids circulation flux, solids feedrates, riser diameter, gas flowrates, aeration rates and the working bed temperatures must be pointed out. The importance of the ratio N_{pass} as a key parameter for design and operation is underlined.

In addition, the less significant effect of design variable such as riser height and total solid inventory is highlighted, provided they remain within normal boundaries. A comprehensive procedure is proposed for the design of a unit to thermally treat industrial wastes.

4.7 Acknowledgement

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CONCLUSIONS

Le Lit Fluidisé Circulant Interne présente une technologie attrayante pour le traitement thermique de déchets industriels sur site, ce qui est une façon efficace de réduire les coûts tout en satisfaisant aux exigences environnementales. La performance de cette unité est fonction des différents paramètres opératoires et géométriques. Un design efficace et une stratégie d'opération adéquate sont nécessaires pour atteindre de bonnes performances. Le présent travail a pour objectif la modélisation et la simulation du LFCI en vue de proposer des critères et des stratégies utiles pour le design et l'opération de cette unité. La modélisation mathématique, telle que proposée dans ce travail, permet de décrire et de caractériser les paramètres importants pour le traitement thermique de déchets industriels à l'aide du Lit Fluidisé Circulant Interne. Cette modélisation est basée sur les phénomènes d'échanges du système et permet d'étudier la performance du réacteur opérant en régime permanent ou transitoire.

Les paramètres d'entrée pour le modèle sont : les débits de gaz et du déchet solide, le taux de circulation du solide, les dimensions du réacteur, et les propriétés physiques des différentes phases. Ces paramètres ne sont pas totalement indépendants car ils sont couplés par l'hydrodynamique, le transfert de chaleur et de masse dans le LFCI. La simulation permet de prédire les propriétés hydrodynamiques, la température, la conversion du déchet et les concentrations des émissions gazeuses.

L'étape initiale de la modélisation consiste à utiliser des hypothèses et des simplifications justifiables. Les équations descrivant l'hydrodynamique, le transfert de masse et chaleurs sont développées en faisant usage de certaines correlations et modèles trouvés dans la litérature. Ces équations sont couplés à la cinétique d'un système réactionnel très simple pour prédire les performances du réacteur, notamment : la conversion, la température, le temps de séjours et la porosité dans le tube de monté. Les prédictions du modèle et les résultats expérimentaux se comparent d'une manière satisfaisante pour la période de préchauffage du sable propre et le traitement du déchet. Ce modèle est particulièrement applicable au traitement de déchets à faible pouvoir calorique dont le traitement thermique nécessite l'usage d'un combustible supplémentaire. Lorsque les émissions gazeuses ne sont pas importants, cette première phase du modèle est suffisante.

Dans la seconde étape, le modèle est modifié de façon à pouvoir calculer les concentrations des émissions gazeuses. Après analyse de différentes données trouvées dans la littérature, un système réactionnel est déterminé en utilisant une série de réactions homogènes et hétérogènes. Dans certaines de ces réactions le char et le solide du lit agissent comme catalyseur. Les données expérimentales obtenues dans cette étude ne permettent pas de déterminer directement les cinétiques ou les mécanismes des réactions impliqués. Cependant l'analyse de ces données démontrent clairement l'implication des matières solides du lit dans la formation des émissions gazeuses comme l'oxyde d'azote. La participation du monoxyde de carbone est aussi vérifiée.

Étant donné que l'activité de ces composés varie selon leur source de provenance, un facteur d'activité est utilisé pour prédire la tendance observée dans les concentrations des émissions gazeuses. Le modèle peut donc être utilisé pratiquement pour une estimation rapide des concentrations du CO, NOx, N_2O, SO_2,O_2 et CO_2 ainsi que pour une estimation préliminaire des paramètres opératoires.

L'un des objectifs principaux de la modélisation est de développer des critères et stratégies utiles pour le design et l'opération de l'unité commerciale. Le LFCI est utilisable comme unité mobile et est capable de traiter les déchets de différentes sources. Sa performance est beaucoup fonction des conditions opératoires. Ce modèle s'applique bien lorsque les températures d'opération sont au-dessus de 850~900°C où le fonctionnement de l'unité correspond aux hypothèses qui régissent le modèle.

Le modèle montre sa capacité d'adaptation à la variation des propriétés du déchet telles que l'humidité, la taille des particules et la composition chimique. Les simulations obtenues pour le traitement des sols contaminés et des boues sont pratiquement en accord avec les données expérimentales. De plus, l'analyse fait ressortir l'importance du rapport N_{pass} comme paramètre clé pour le design et l'opération. Par contre, la hauteur du riser et la masse totale du lit ont un effet limité. La procédure de design proposée dans cette étude contient de l'information pratique pour le développement d'une unité industrielle.

RECOMMANDATIONS

Cette étude propose un modèle mathématique prédictif pour le traitement de déchets industriels et montre aussi indirectement la faisabilité d'un tel procédé. Le modèle utilise certaines données caractéristiques du déchet et de l'unité comme le coefficient de transfert de chaleur pour les pertes thermiques et la cinétique de combustion du déchet. Les données cinétiques provenant directement de la littérature possèdent certaines limitations, principalement pour les réactions hétérogènes catalytiques auxquelles il a fallut utiliser un facteur de correction. Il serait souhaitable de développer des cinétiques propre au déchet et relatives à la génération des émissions gazeuses.

D'autre part, le modèle hydrodynamique est relativement très simple mais très pratique aussi. L'obtention des propriétés hydrodynamique est difficile pour les opérations à haute température. Ce qui fait que sa validation a été faite uniquement avec des données limitées. Il serait bon de le faire aussi avec des données se rapportant à la porosité et à la vitesse des particules dans le tube de montée. Toutefois, l'effet de la circulation du solide minimise l'erreur introduite par l'usage d'un modèle simple pour le tube de montée.

Pour une modélisation beaucoup plus complète, il est suggéré de chercher le moyen de prédire aussi le taux de circulation du solide en fonction des vitesses superficielles du gaz dans le tube de montée et l'annulaire, et des trous situés à la base du tube de montée. Ceci permettrait d'éviter d'utiliser le taux de circulation comme paramètre d'entrée.

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ANNEXE A : Données Techniques et mesures

Unité expérimentale

L'unité expérimentale est présentée à la figure 1. Le bas du tube de montée est fait de façon à modifier la taille des trous permettant la circulation de solide annulaire à travers le tube de montée. Dans cette étude, une bague ayant 8 trous de 1 cm chaque a été utilisée.

Brûleur contre-rotation

Ce brûleur, Figure A-1, est basé sur la mise en rotation en sens inverses de l'air comburant sur deux étapes d'ouvertures tangentielles. Cette approche favorise la turbulence et le mélange de l'air avec le gaz qui est injecté radialement à la base du brûleur. La flamme de ce type de brûleur a la caractéristique d'être dense, compacte et très stable.

Le brûleur a une puissance nominale de 20 kW. Il est muni d'une électrode de détection de flamme et une bougie d'allumage. l'électrode de détection est couplée à un relais responsable de fermer l'électrovanne qui coupe le débit de gaz lorsque l'électrode ne détecte plus de flamme.

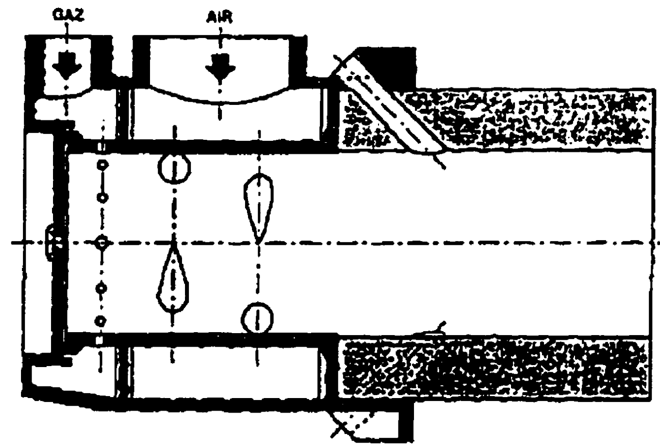


Figure A-1. Vue schématique du brûleur à contre-rotation.

Températures

Les thermocouples de type B permettent de mesurer les températures situées entre 400~1800°C. Il servent à prendre les températures dans le tube de montée. Les thermocouples de type K sont appropriés pour des températures de -40~1100°C, ils sont utilisés pour mesurer les températures dans l'annulaire, la zone de désengagement, les fumées de combustion, la paroi externe du lit et la température ambiante.

Les températures à l'intérieur du tube de montée sont le résultat de la conduction, la convection et la radiation créée par le gaz, le solide et les parois. Des gaines en céramique protègent le thermocouple de type B contre les frictions produites par l'écoulement gaz-solide tout en réduisant la quantité de chaleur transmise au thermocouple par radiation. La présence des particules solides génère de grands coefficients de transfert de chaleur de telle sorte que la température mesurée se rapproche beaucoup de la température du mélange gaz-solide.

Débits de gaz et capteurs de pression

Les débits d'air primaires, secondaires, pneumatiques et de gaz naturel sont mesurés à l'aide de débitmètres à orifice installés sur les trains respectifs. La différence de pression est mesurée par des capteurs de pression à diaphragme qui fournissent un signal électronique en milliampères. Ce signal est transformé en volts à l'aide d'une résistance avant d'être acheminé et interprété par une carte d'acquisition de données. Le signal en volts est proportionnel au débit. Les données respectives sont présentées au tableau AI-1.

L'erreur systématique sur les mesures de débits gazeux dépend de l'incertitude des différentes composantes : l'orifice en lui-même (2~3%), les capteurs (0.25~1%, selon le modèle), la résistance transformant le signal et la carte d'acquisition de données. L'analyse des fumées de combustion et l'usage de compteur à gaz ont été mis à contribution pour confirmer la validité des débits. Globalement l'erreur est moins de 5%.

Tableau A-1. Principales données techniques des capteurs et débitmètres.

	Air principal	Air secondaire	Gaz naturel	Riser
Diamètre de l'orifice (cm)	1.067	1.00	0.8293	7.8
Capteurs de pression respectifs				
modèle	Modus R-32	Omega PX272-10DI	Omega PX272-01DI	Validyne P-306
ΔP_{max} (kPa)	24.91	2.49	0.25	0.25
voltage	24	24	24	24
Sortie(mA)	4.2	4.2	4.2	4.2
Précision-sortie (pleine échelle)	0.5%	1%	1%	0.25%

Débit de solide

Le débit de solide est proportionnel à la vitesse de rotation du moteur de la vis d'Archimède servant à alimenter le solide. Ce débit a été calibré en fonction de la lecture du potentiomètre contrôlant la rotation de la vis. La courbe de calibration est présentée à la Figure A-2, ci-dessous :

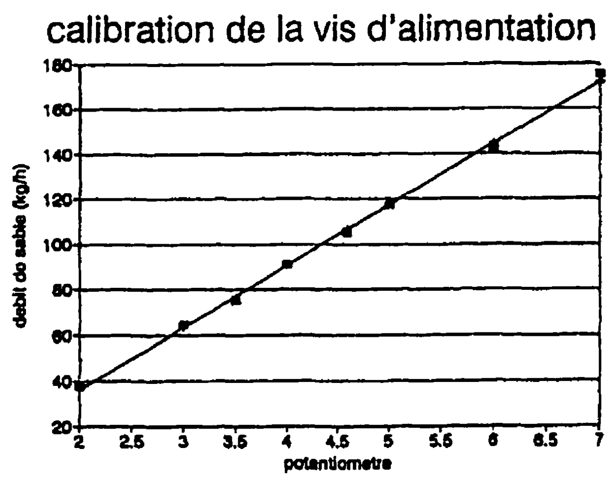


Figure A-2. Courbe de calibration de la vis d'Archimède.

Concentration et conversion de la résine organique

La concentration de la résine est déterminée en réalisant une perte au feu à 900~950°C pendant près de 3 heures. La différence de poids avant et après la perte au feu permet de calculer la quantité de résine dans l'échantillon. Le processus est résumé comme suit :

- peser un creuset vide et sec,
- peser le creuset rempli d'un échantillon de sable,
- placer l'échantillon et le creuset au four à 900~950°C durant 3 heures,
- retirer le creuset puis laisser refroidir, 3 heures,
- peser le creuset et son contenu,
- calculer le pourcentage du poids perdu par l'échantillon, représentant ainsi le pourcentage de résine.

Taux de circulation

Le traçage thermique est utilisé pour mesurer le taux de circulation. Il consiste à injecter près de 5 kg de sable froid à travers l'annulaire. Le passage de ce sable froid produit des signaux thermiques détectés par les thermocouples placés verticalement dans l'annulaire. Trois thermocouples baignent normalement dans le lit de sable annulaire lorsque la masse du lit est de 120~140 kg. La distance entre ces thermocouples est de 45 cm. L'évolution de la température en fonction du temps génère des pics comme à la Figure AI-3. Le temps qui sépare deux piques sert à calculer la vitesse de descente du lit et en connaissant la densité en vrac (ρ_{bulk}) ainsi que les sections respectives de l'annulaire et du riser, le taux de circulation est estimé comme suit :

$$\text{Taux de circulation, Gs} = \frac{\text{distance (45cm)}}{\text{temps écoulé}(\Delta t)} \rho_{bulk} \frac{A_{annulaire}}{A_{tube\ de\ montée}}$$

La faible variation de la variance et de la température moyenne avec la position démontre la fiabilité de cette méthode. La chaleur reçue par le sable durant le passage n'introduit pas d'erreur considérable.

Le taux de circulation est directement relié à la chute de pression dans le tube de montée. Pour une vitesse superficielle du gaz et un taux de circulation fixes correspond aussi une chute de pression. Les chutes de pression observée se comparent d'une manière satisfaisante avec d'autres valeurs trouvées dans la littérature, voir chapitre 2.

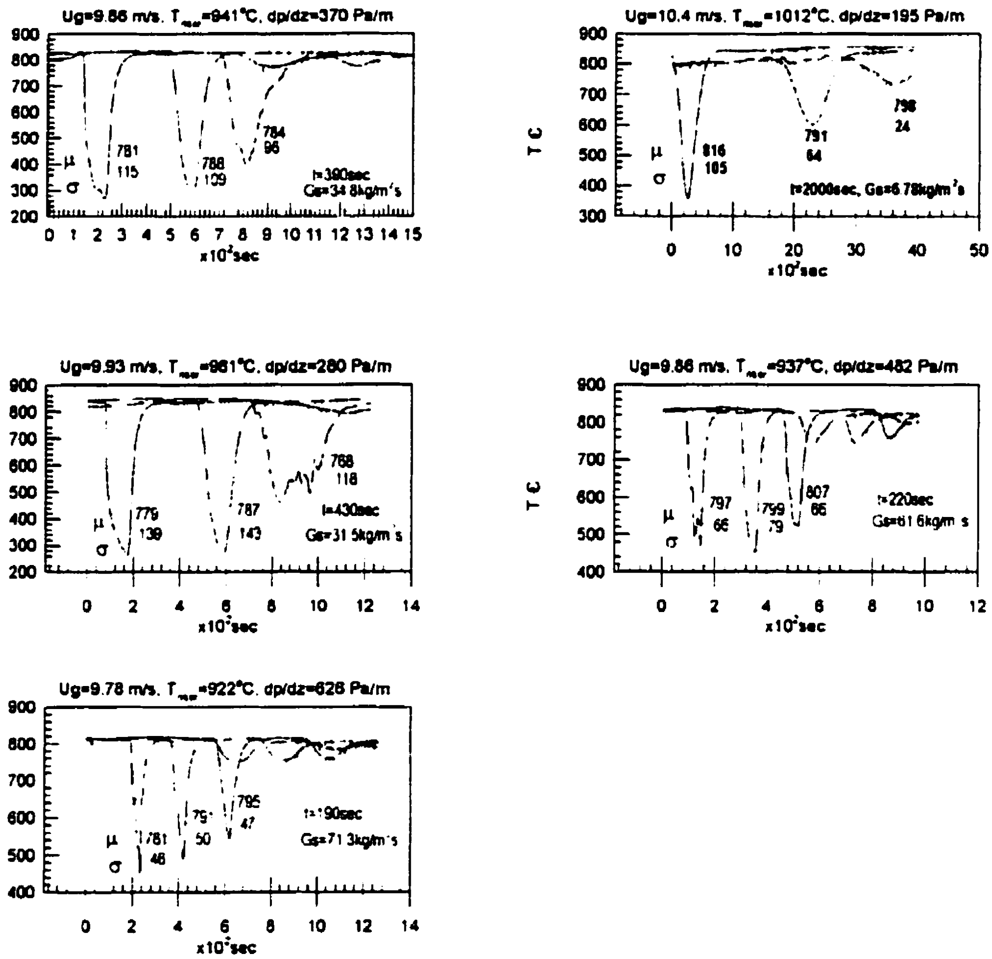


Figure A-3. Essai pour la mesure de taux de circulation par traçage thrmique.

Analyse des fumées de combustion

Un analyseur de fumée de marque « Eurotron » et de modèle « Greeline-Combustion gas analyser » est utilisé pour mesurer les concentrations volumiques de O_2, CO, NO_x et SO_2 à l'aide de cellules électroniques indépendantes. L'analyseur calcule la concentration de CO_2 par bilan à l'aide de l'équation chimique sur la base du choix du combustible; ces resultats sont donc erronés puisque non seulement non le gaz naturel mais aussi la matière organique du déchet brûle. Les concentrations d'imbrûlés sont déterminées à l'aide d'un analyseur à infrarouge. Le tableau AI-2 présente l'échelle de mesure, la résolution et l'erreur systématique pour les concentrations mesurées par ces appareils.

Tableau A-2. Échelle de mesure, résolution et erreur systématique pour les concentrations mesurées (concentrations volumiques)

Composé	Échelle	Résolution	Erreur systématique
O_2	0~25%	0.1 %	<1 %
CO	0~4000 ppm	1 ppm	< 4%
NOx	0~4000 ppm	1 ppm	<4 %
SO_2	0~2000 ppm	1 ppm	<4%

ANNEXE B : La thermogravimétrie

La thermogravimétrie a été utilisée pour obtenir la cinétique de combustion du sable de fonderie. Cette technique est présentée au chapitre 2 qui donne aussi certaines références pertinente. La technique consiste essentiellement à mesurer la perte du poids du solide en fonction du temps pendant que la combustion a lieu. La vitesse de chauffage, les débits de gaz et les concentrations doivent être fixés. La Figure B-1 montre le résultat d'un essai d'analyse thermogravimétrique. Les résultats produits sous différentes conditions sont regroupés à la figure B-2.

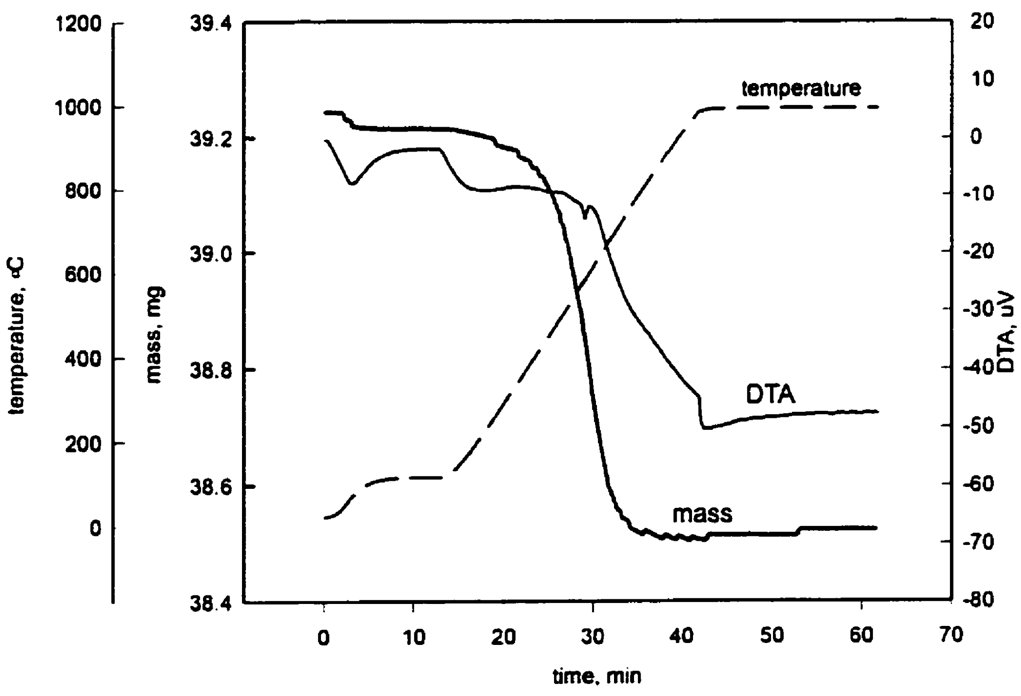


Figure B-1. Résultats d'un essai d'analyse thermogravimétrique

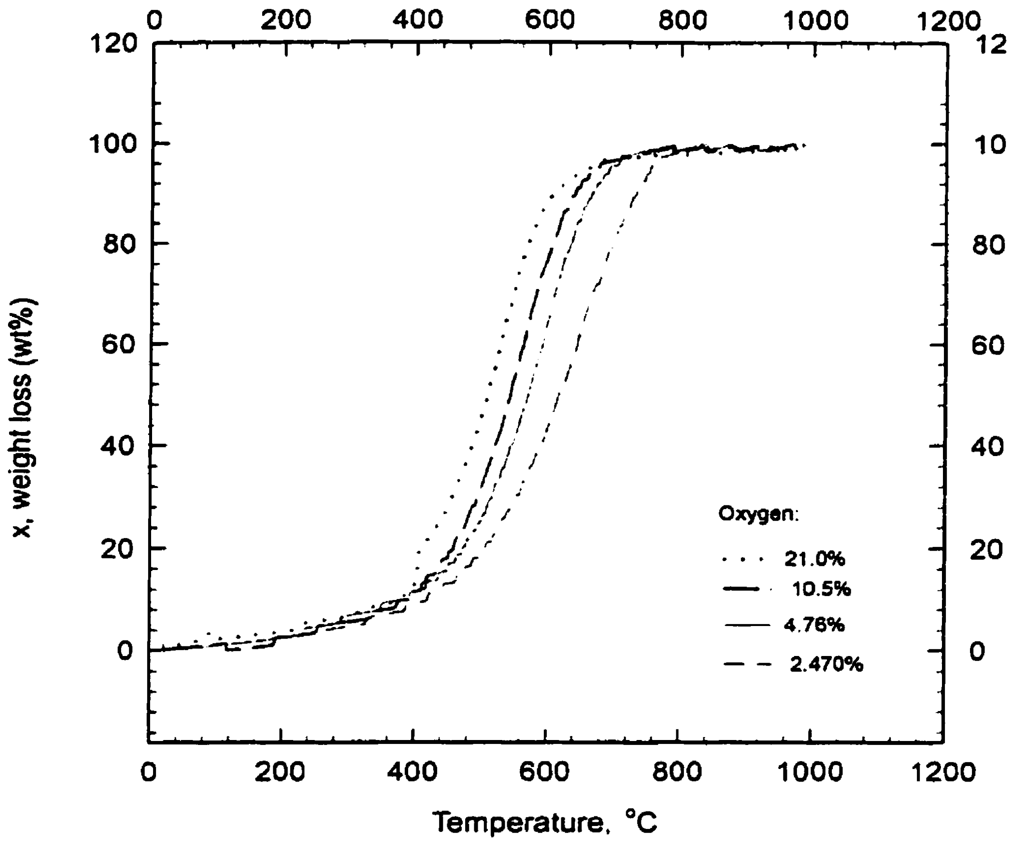


Figure B-2: Perte de mass en fonction de la temperature et de la concentration d'oxygène pour un taux de chauffage de 30°C/min.

ANNEXE C : Données Expérimentales Sans Lit de sable

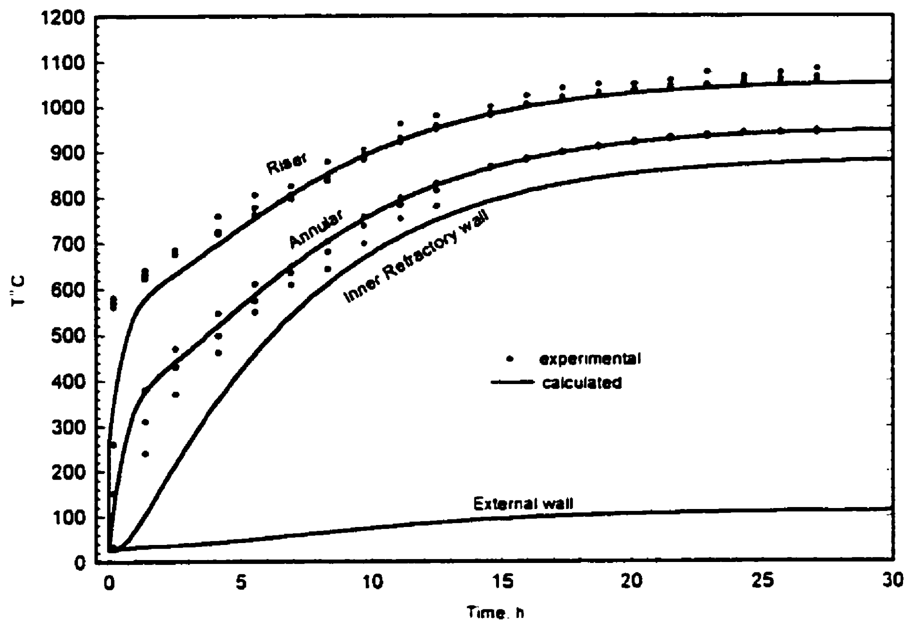
Émissions gazeuses

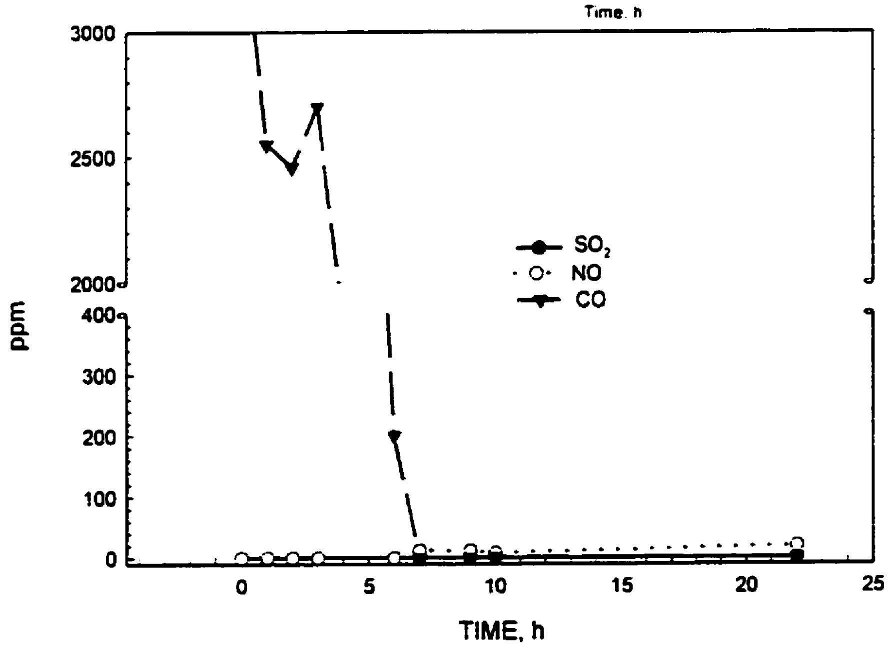
T	Air	aération	O2	CO	CO2	NO	SO2
1120.	21.8	1.09	1.90	1.00	9.50	86.0	2.00
1110.	25.0	1.24	4.40	1.00	9.30	47.0	0.00
980.	29.0	1.43	6.10	0.00	7.10	26.0	0.00
1040.	35.0	1.73	9.30	15.0	5.80	8.00	0.00
900.	40.0	2.12	11.7	275.	4.60	0.00	0.00
1120.	21.0	1.04	0.70	19.0	10.2	87.0	2.00

ANNEXE D : Données Expérimentales avec Lit de sable propre

Profil de température ,et émission gazeuses:

T_{room}=35°C, $m_{s,tot}$=140 kg, W_{air}=37.7 kg/h, aeration=1.4, Gs=35 kg/m²s, ws=0 kg/h, $k_{blanket}$ = k(T) W/m°C, k_{sand}=1.82 W/m°C, $h_{w,o}$=13 W/m² °C

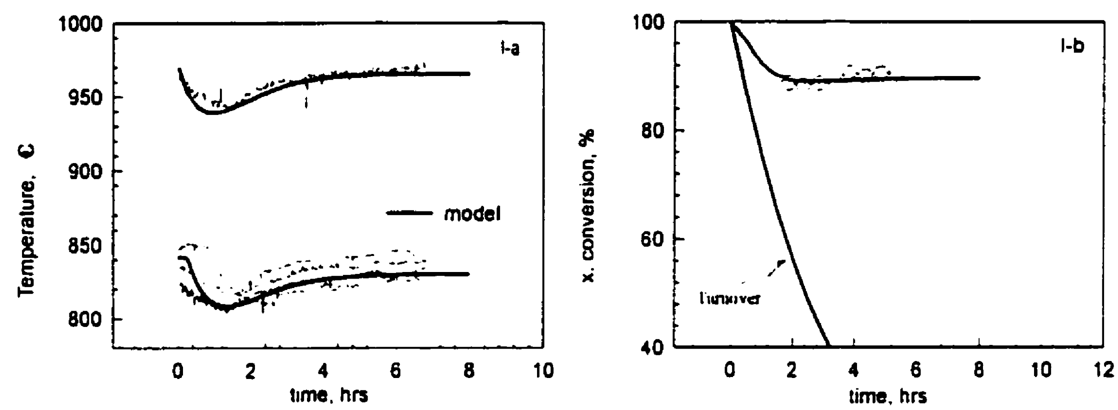


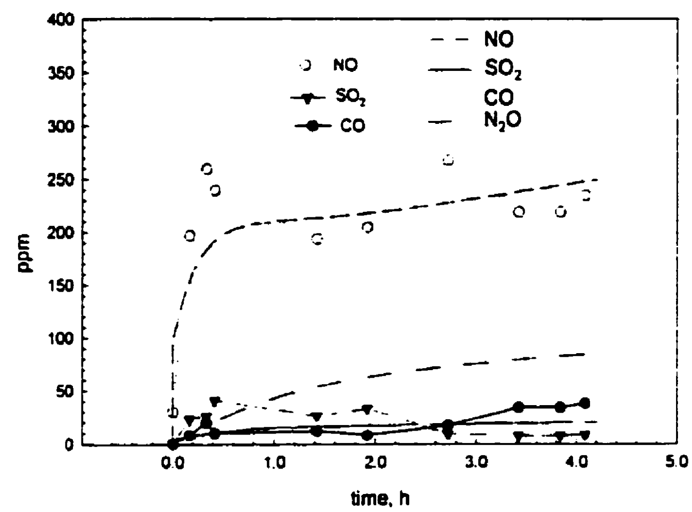


ANNEXE E : Données Expériementales avec Sable Usé

ESSAI- 1: Profil de température et Émissions gazeuses:

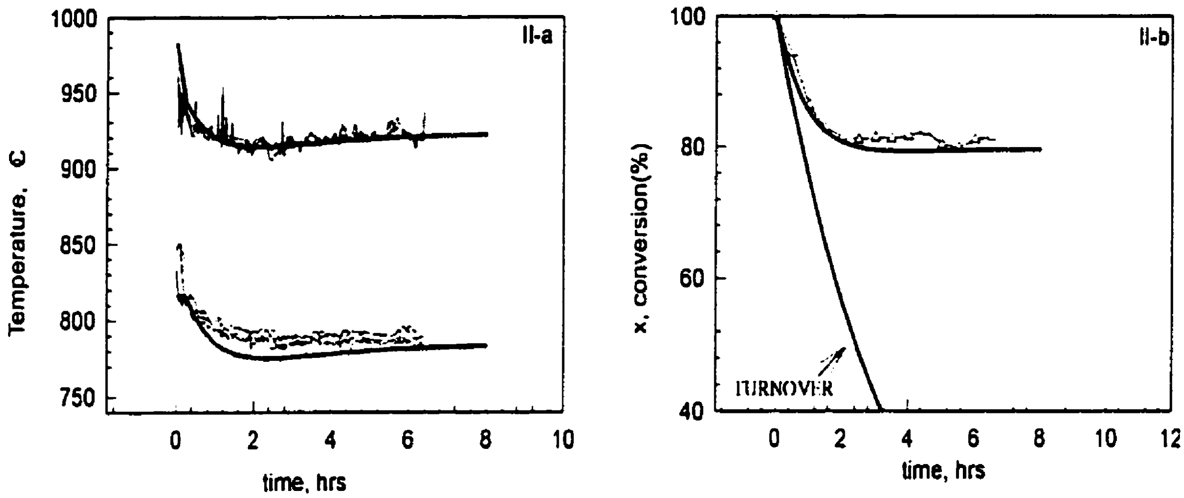
W_{air}=37.7 kg/h, aeration=1.35, W_{feed}=35 kg/h, $m_{s,tot}$=100 kg, Gs=35 kg/m^{2}s, from room T=35°C, $h_{w,o}$=13 W/m^2 °C, $h_{w,i}$=32 W/m^2 °C, $k_{blanket}$=k(T) W/m°C.

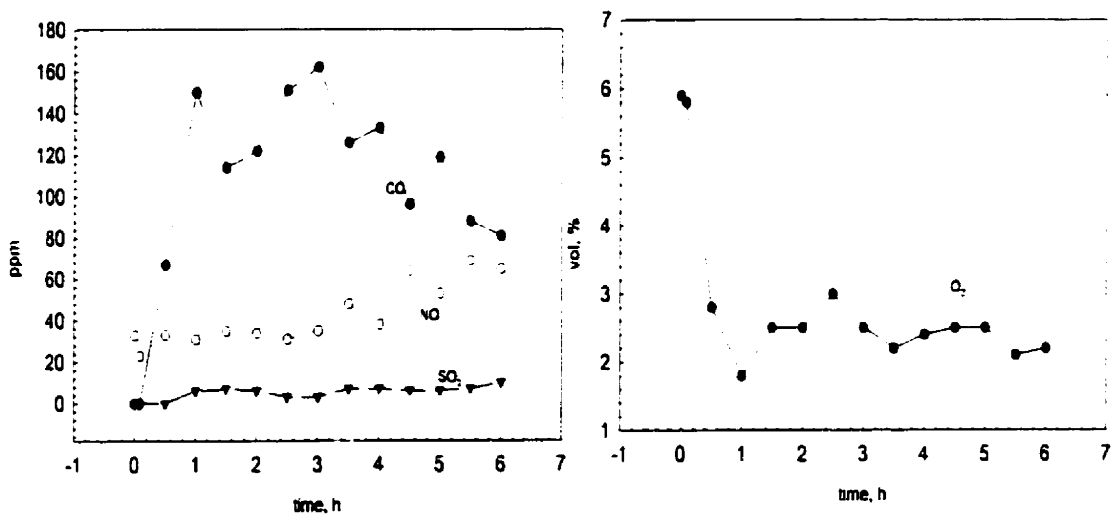




ESSAI- 2: **Profil de température et Emissions gazeuses:**

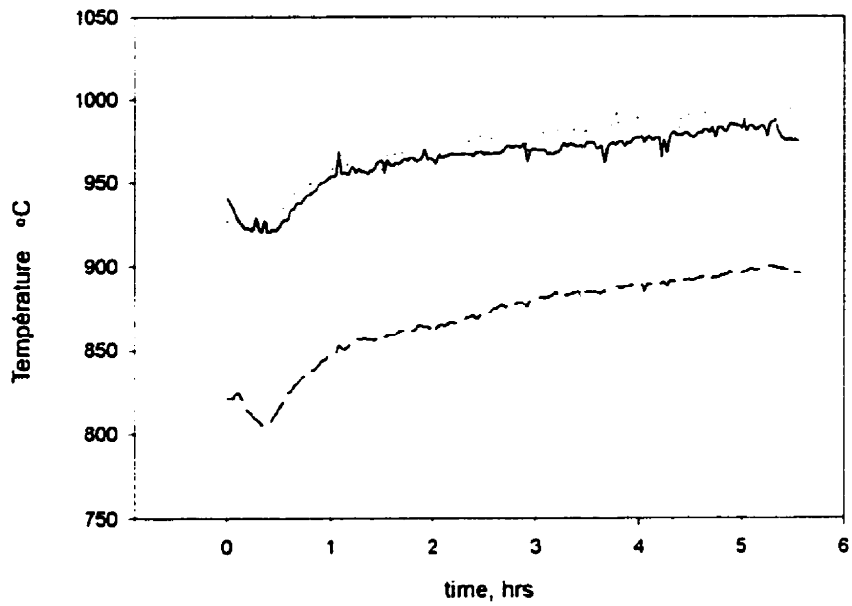
W_{air}=34.8 kg/h, aeration=1.2, W_{feed}=41 kg/h, $m_{s,tot}$=140 kg, Gs=50 kg/m^{2}s, from room T=15°C, $h_{w,o}$=13 W/m^2 °C, $h_{w,i}$=33 W/m^2 °C, $k_{blanket}$=k(T) W/m°C.

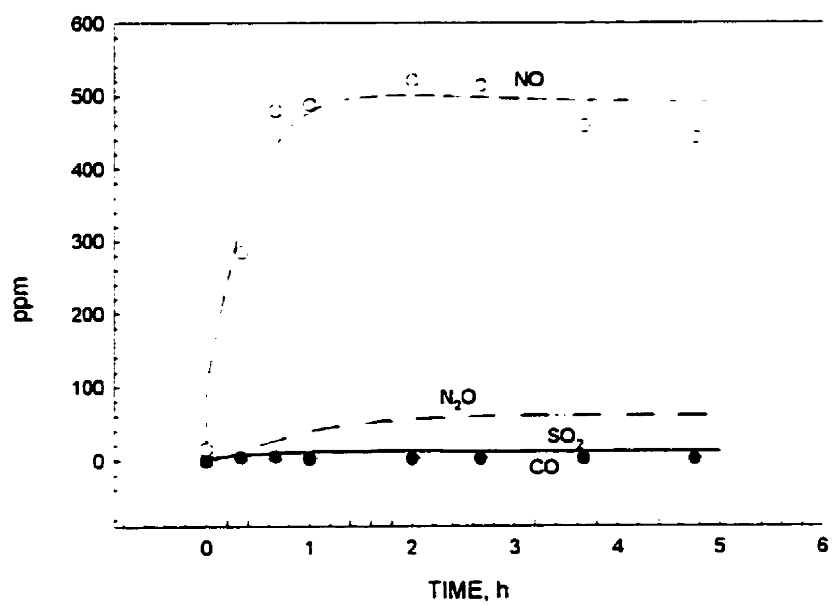




ESSAI- 3: Profil de température et émissions gazeuses:

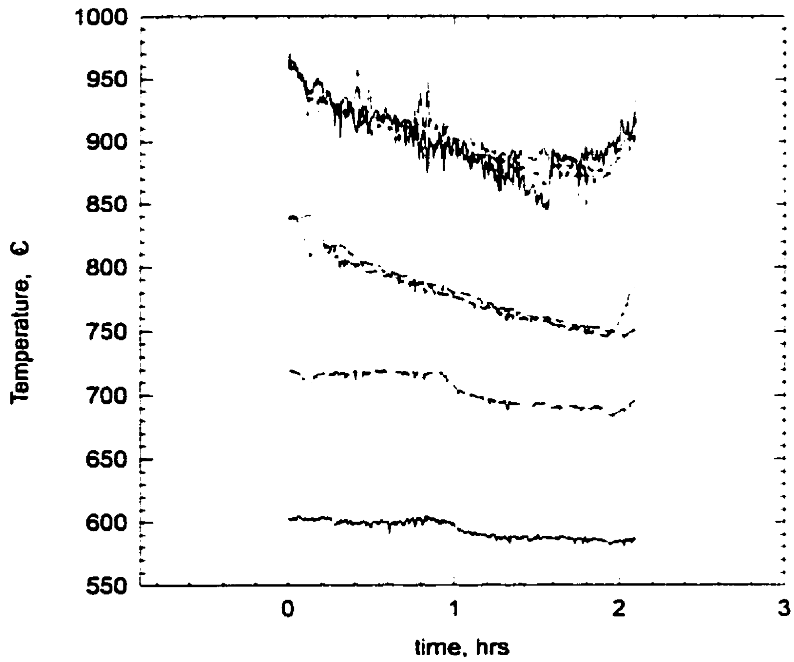
Débit solide: 20 kg/h; Gs: 37 kg/m²s, masse$_{lit}$=140 kg (initiale), X $_{conv}$> 95%





ESSAI- 4: Profil de température et Emissions gazeuses:

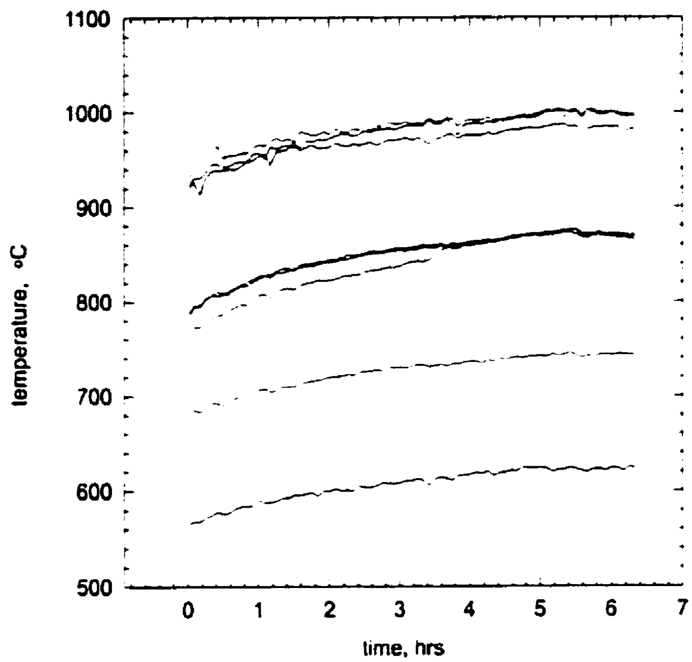
Débit solide: 60 kg/h; Gs: 48 kg/m^{2}s



T°C	T_{Chem}	air	n	O_2	CO	NO	SO_2	time	X
920.	236.	29.	1.47	6.5	0.	29.	0.	0.	100
	236.	29	1.47	3.8	169.	30.	11.	0.33	94
	225.	29	1.47	4.9	87.	55.	21.	0.83	87
	239.	29	1.47	5.3	102.	135.	28.	1.33	84
	245.	29	1.47	5.4	182.	46.	16.	1.83	82

ESSAI- 5: Profil de température et Emissions gazeuses:

Débit solide: 20 kg/h; Gs: 35 kg/m^{2}s, masse$_{lit}$=70 kg (initiale), X_{final}>95%

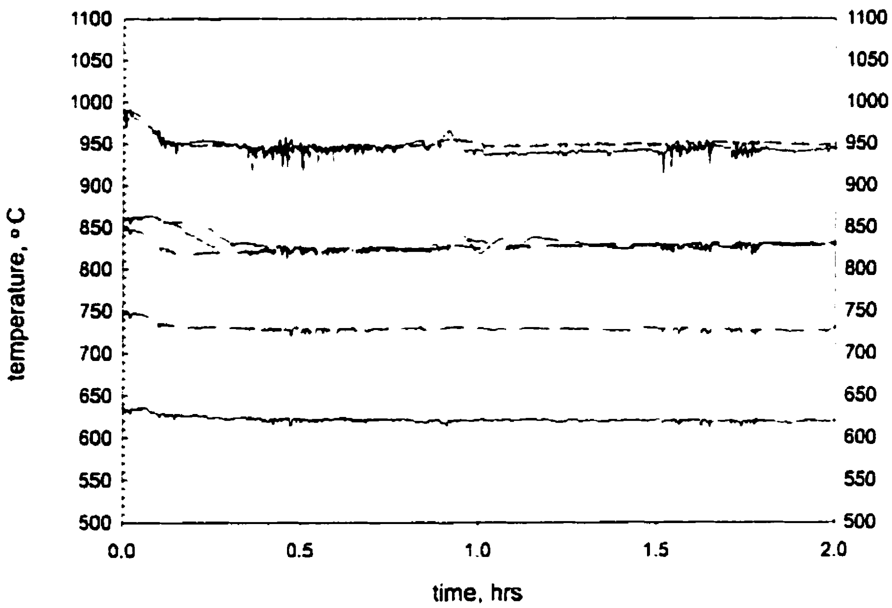


T°C	T_{Chem}	air	n	O_2	CO	NO	SO_2	time
960.	208.	28	1.47	6.3	3.00	230.	4.0	-1.00
960.	208.	28.	1.47	6.0	8.00	330.	0.0	0.00
968.	223.	28.	1.47	4.9	5.00	275.	1.0	2.70
975.	234.	28.	1.47	4.3	2.00	294.	0.0	4.05
985.	202.	28.	1.47	4.2	0.00	255.	5.0	5.75

ESSAI- 6: Profil de température et Emissions gazeuses:

Débit solide: 40 kg/h; Gs: 35 kg/m^{2}s, masse$_{lit}$=140 kg (initiale),

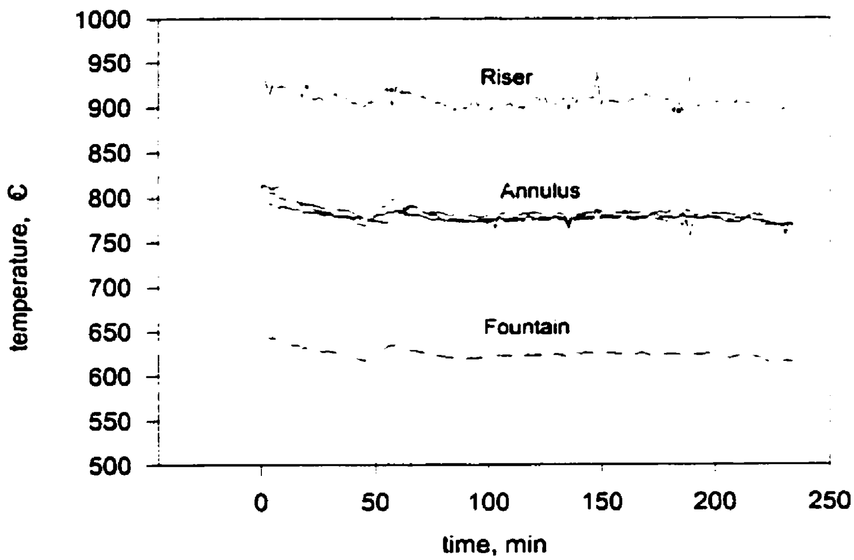
essai préliminaire., X_{final} >78%



T°C	T_{Chem}	air	n	O_2	CO	NO	SO_2	time
950.	206.	28	1.43	6.4	7.00	70	1.0	0.0
950.	202.	28	1.43	4.8	18.00	62	1.0	0.17
950.	191.	28	1.43	4.2	24.00	54	5.0	1.0
950.	209.	28	1.43	4.1	24.00	63	3.0	2.0

ESSAI- 7: Profil de température et Emissions gazeuses:

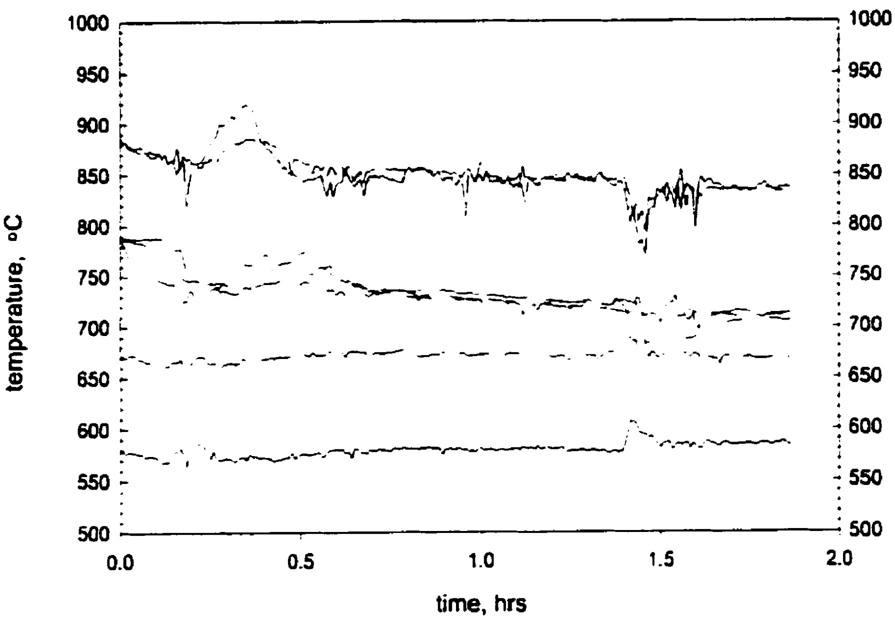
Débit solide: 50 kg/h (sable à noyau); Gs: 40 kg/m^{2}s, masse$_{lit}$=140 kg (initiale)



T°C	T_{Chem}	air	n	O_2	CO	NO	SO_2	time(h)	X%
940		28	1.47	6.3	0	2	0	0.00	100
900.	237.	28.	1.47	3.2	60.	75.	0.0	0.33	
	241.			2.9	16.	99.	0.0	0.67	95.4$_{(t=0.5)}$
	249.			2.7	106.	135.	0.0	1.00	86.4
	255.			3.5	122.	89.	0.0	1.33	85.6
	252.			3.0	14.	71.	0.0	1.67	85.0
	251.			2.2	50.	87.	2.0	2.00	85.7
	249.			2.0	407.	63.	2.0	2.33	85.4
	254.			2.5	24.	31.	3.0	2.67	85.0
	250.			1.6	1.	26.	2.0	3.00	85.0
	250.			1.7	5.	28.	2.0	3.33	84.1

ESSAI- 8: Profil de température et émissions gazeuses

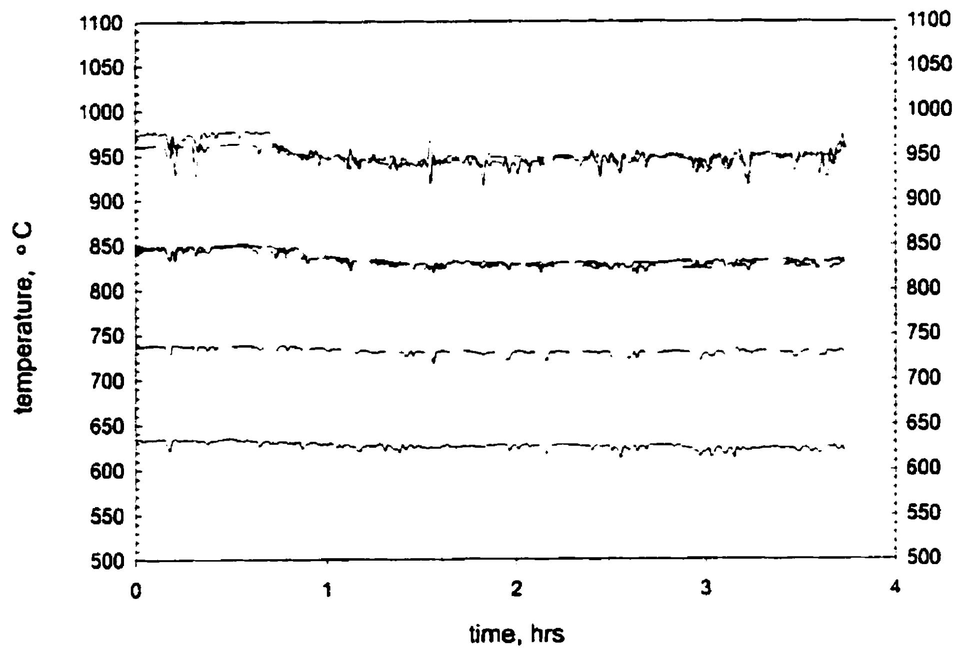
Débit solide: 60 kg/h; Gs: 38 kg/m²s, $masse_{lit}$=140 kg (initiale)



T°C	T_{Chem}	air	n	O_2	CO	NO	SO_2	time	X%
920.	230.	28.	1.43	6.5	0.0	0.	0.		100
920.	201.			6.5	0.0	0.	0.	0.0	100
850.	201.			5.00	22.	31.	0.	1.50	$86.2_{(t=1)}$
835.	197.			4.70	1082.	34.	0.	2.17	$83.8_{(t=2)}$
830.	196.			5.0	970.	27.	0.	2.42	

ESSAI- 9: Profil de température et émissions gazeuses

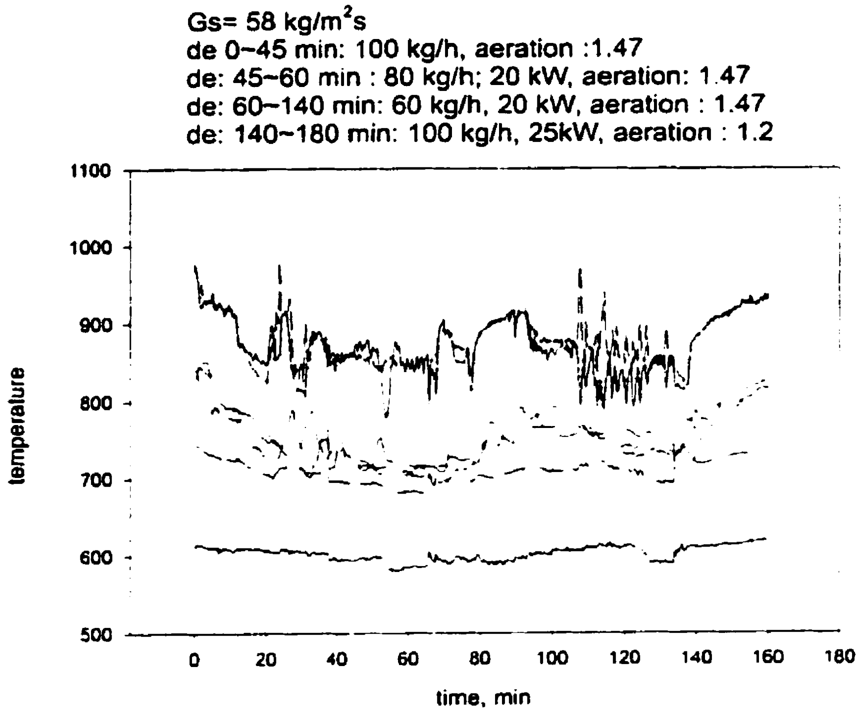
Débit solide: 40 kg/h; Gs: 48 kg/m^{2}s, masse$_{lit}$=140 kg (initiale)



T°C	T_{Chem}	air	n	O_2	CO	NO	SO_2	time	X%
960.	253.	28	1.4	6.2	0.0	16.	0.	0.0	100
950.	225.	28	1.4	10.7	45.	14.	0.	0.75	93.2
950.	249.			2.1	23.	41.	5.	1.0	88.3
950.	249.			2.0	26.	38.	4.	1.5	85.2

ESSAI- 10: **Profil de température et émissions gazeuses**

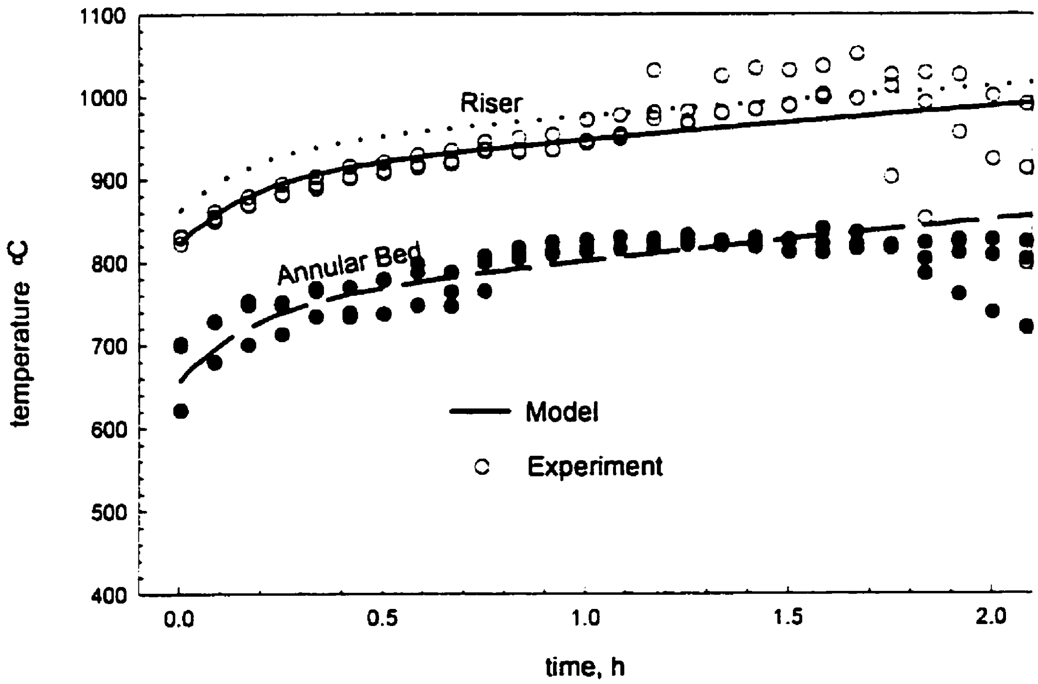
$masse_{lit}$=140 kg (initiale)



Min	O_2	CO	NO	SO_2	position
0	10.5	4500	61	10	entrée Fontaine
30	10.3	4500	146	10	sortie fontaine
45	10.3	4500	160	12	s.f.
60	12.1	4500	67	13	s.f.
72	12.9	4	52	7	cheminée
84	19.8	1353	361	72	riser bas
90	10.4	4500	130	20	rise milieu
98	11.4	4500	425	2	riser milieu
135	9.2	4500	28	9	riser sortie
140	14.3	76	28	5	cheminée
150	6.7	10	41	3	cheminée
160	6.9	67	34	18	entrée fontaine
170	6.6	728	40	0	entrée fontaine

ANNEXE F : le traitement thermique des Boues

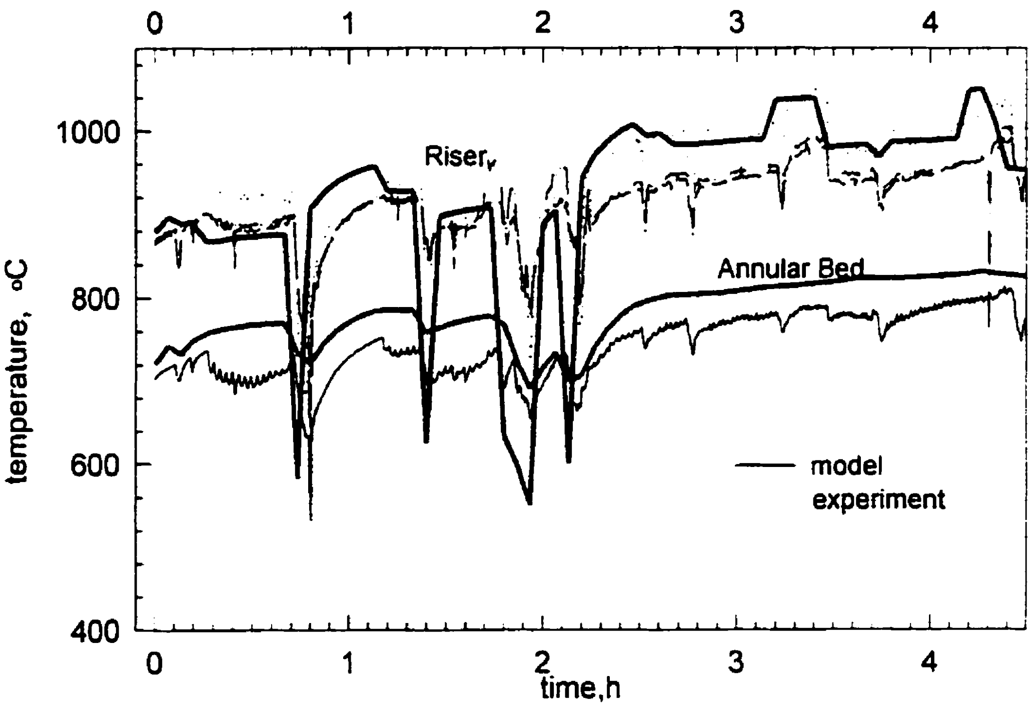
Temperature profile during Deinking Sludge treatment, Air flowrate= 37.4 kg/h; Aeration= 1.87, Initial organic content= 47%; X_{exit}>94%.



ANNEXE G: Traitement des Sols Contaminés

Profil de température:

Comparison between experiment and computed temperature profile for treatment of contaminated soil, $M_{s,tot}$= 100 kg, Gs= 30 kg/m^{2}s, total mass fed: 228 kg.



ANNEXE H: Article publié dans 14th International Coonference on Fluidized Bed Combustion, volume 1, pp627-632, (1997).

DEVELOPMENT OF AN INTERNALLY CIRCULATING FLUIDIZED BED COMBUSTOR FOR TREATMENT OF INDUSTRIAL SOLID WASTES

L. Mukadi, R.-J. Lavallée*, R. Legros and C. Guy*

Gas Technology Research Group, BIOPRO Research Center
Department of Chemical Engineering, École Polytechnique
P.O. Box 6079, st. "Centre-ville", Montreal, Quebec, H3C 3A7, Canada

ABSTRACT

A novel thermal treatment technology for low heating value wastes has been tested at the pilot scale level. The first application deals with reclamation of foundry sand. This waste is produced after several cycles of mold making and the resulting spent foundry sand particles is covered with an organic resin. Because of this resin, the waste is classified special waste and this leads to high landfilling costs, not taking into account the replacement cost of this sand for the foundries.

The internally circulating fluidized bed (ICFB) unit showed excellent performances for treating spent foundry sand. The high temperature contact between the solids and the flame region of a natural gas burner provided high combustion efficiency while, maintaining high overall energy efficiency. Indeed only a small region of the reactor that is the base of the riser is kept at high temperature. The remainder of the unit can be kept at lower temperature, which is not possible in conventional fluidized beds normally used for foundry sand thermal reclamation where the entire bed is maintained at the treatment temperature. The specific energy consumption is therefore very competitive for the ICFB and emission levels were low in CO and NOx. A brief economical assessment of using an ICFB for thermal reclamation of spent foundry sand shows relatively short payback times for a typical foundry.

INTRODUCTION

Many types of industrial wastes generated today require treatment in order to comply with environmental standards. The industrial wastes considered here are organic solid wastes of low heating value which require the use of supplementary fuel to sustain combustion, such as:

- Pulp and paper deinking sludge
- Other divided solid wastes
- Wastewater treatment sludge
- Electrodes from the aluminum industry
- Contaminated soil
- Industrial sludge
- Spent foundry sand

A novel type of fluidized bed reactor, the Internally Circulating Fluidized Bed (ICFB), has been developed at the Gas Technology Research Group of Ecole Polytechnique of Montreal to treat such wastes. As with any new processes, technological and economical feasibility must be demonstrated.

For this purpose, spent foundry sand has been selected to evaluate the new technology at the pilot scale level and to set grounds for further industrial applications. Spent foundry sand

* presently with Marc Dufresne inc., Trois-Rivières-Ouest, Quebec
* author for correspondence

disposal is capturing the attention of several foundries [Reier 1993, Lessiter 1994, Philbin 1995, Bralower 1989].

Foundry sand - Background:

Foundry sand is the result of mixing clean sand with a binder. It is then used to fabricate molds for metal casting. Foundry sand may be used several times to make molds for metal casting, after which it loses its molding capacity and become spent foundry sand. The latter is coated with a degraded binder and must be handled by appropriate waste disposal methods. A typical foundry in Canada generates 1 000–10 000 tons of spent foundry sand per year. There are two major methods for the management of spent foundry sand: landfill disposal and reuse after reclamation. The increasing costs of landfilling, the rising costs of clean sand and the severity of environmental regulations, make landfill disposal less and less attractive. As a result, foundry sand reclamation starts to make economical sense as well as environmental sense. This has led to the development of several reclamation technologies.

Foundry sand reclamation processes involve physical, chemical or thermal treatment of the sand with a single purpose which is to remove the degraded binder without significantly lowering the original molding properties of the sand. Despite the numerous existing reclamation technologies, a typical foundry sand reclamation involves four major steps:

1. the removal of the casting from the mold by shakeout and crushing;
2. the breakup of the mold to manageable grain size;
3. the reclamation treatment to remove the maximum amount of the residual binder from the grain without fracture of the grain;
4. the restoration of the original sand size distribution.

Steps 1 and 2 are physical processes performed by standard equipment. Most reclamation processes differ in the accomplishment of step 3 and 4. In principle, there are 3 basic systems of reclamation:

- Wet reclamation
- Mechanical reclamation
- Thermal reclamation

Wet reclamation uses water to dissolve water-based binder such as sodium silicates and clays. The spent sand is vigorously agitated within a bath of water to dissolve the binder. Once the sand leaves the water, it must be dried. The drying require a lot of energy which makes it expensive. Also wet reclamation ... of high pH water which creates

Again, mechanical reclamation creates fines, dust and residuals that must be captured in a dust collector, therefore generating another solid waste.

Thermal reclamation entails the heating of the grains to the point of oxidation to combust the organic coating. In order to sustain combustion of the low heating value resin, additional energy input is required, in the form of gas or electricity. Thermal reclamation generates flue gases containing fines as a waste stream.

Thermal reclamation of foundry sand:

The choice of a treatment process depends on the physical and the chemical characteristics of the waste to be treated. Thermal reclamation is mostly effective for spent foundry sand coated with organic binders such as phenolic resins. Several processes of thermal reclamation have been developed and they differ in their configuration, heating method and residence time of the grain within the heating chamber. Fluidized beds and rotary kilns are two systems that are widely used for the reclamation of foundry sand [Lessiter 1994, Lumsden 1993, Philbin 1995]. In fluidized beds, the flowing gas creates turbulence within the suspended mass of solids, providing good mixing and uniform particle heatup. Rotary kilns rely on the mechanical rotation of the kiln to achieve mixing for uniform particle heatup. They require higher excess air ratios than fluidized beds to achieve comparable combustion efficiency.

At the Gas Technology Research Group of Ecole Polytechnique of Montreal, a gas-contact process was developed in which gases and spent foundry sand flow downward in a type of downflow reactor [Benali et al.,1992]. In this type of reactor, the solid phase is directly injected into the flame of a natural gas burner, which provides a high temperature treatment zone. The residence time and conversion were not easy to control and were found to depend highly on the length of the combustion chamber, since particles flow by gravity. It was found that the technology could not treat wastes that require longer residence time such as large particles, highly concentrated waste or humid waste. A novel type of reactor is proposed, the ICFB, which combines all the advantages of fluidized beds and the high temperature solid treatment provided by the gas-contact process. Solids residence time can be controlled, together with higher temperature and high mixing of phases. The originality of this gas-contact process is assured by the presence of the burner which enables direct waste-flame contact propitious for rapid devolatilization and fast combustion of wastes. The technical and economical feasibility of this process are presented in this paper.

1991]. New applications extend its uses as a catalytic reactor [Pugsley et al 1992] and a combustor for industrial treatment of wastes [Sadhukhan &Bradford 1993, Wilbourn et al 1986, Mullen 1992]. There are many published theoretical and experimental studies for the classical CFB which is composed of a riser, a cyclone and an external downcomer as in Figure 1. The ICFB includes three hydrodynamically distinct zones that are similar to those of a conventional CFB: the riser, the disengagement zone or fountain, and the annulus. In addition, the ICFB incorporates the characteristics of a CFB and a spouted bed [Milne et al., 1992].

The present reactor is composed of two concentric cylinders and a natural gas burner vertically located at the base of the inner tube, or riser, in such a way that the flame develops in the riser. The riser is shorter than the outer tube. Located at the end of the riser is the solid disengagement zone or fountain. Particles from the disengagement zone fall into the annular space where they undergo downward flow. Secondary air is used at the base of the outer tube to control the solid circulation rate. Solids from the annular space re-enter the riser through perforated holes at the riser bottom. In the present application, the spent foundry sand is fed through the burner, though solids can also be fed directly into the annular region. An US patent application has been filed for this technology (Guy et al., 1996).

The solid circulation rate, Gs, is an important design parameter for both the modeling and the operation. It influences the residence time of particles in the annulus bed, the solids holdup in the riser and the temperature profile in the reactor. At higher Gs, solid phase behavior in the reactor will approach complete mixing (CSTR).

EXPERIMENTAL

Experimental Procedure:

An ICFB pilot scale reactor was constructed and installed at the Gas Technology Research Group of Ecole Polytechnique of Montreal (see Figure 2). The reactor is made of two concentric stainless steel cylinders (riser and external cylinder), a cyclone and a natural gas burner. The inner walls of the external cylinder (SS 304) are covered with a 3 cm layer of refractory and a 2.54 cm layer of ceramic wool. The riser is 1.5 m in height, 7.8 cm inner diameter and 8.9 cm outer diameter. The external cylinder is 2.5 m in height and 35 cm inner diameter with the refractory.

The solid feed-system is composed of a screw-conveyor and pneumatic transport section. The screw-conveyor regulates the solid feed rate. Pneumatic conveying is used to transport the solid into the reactor. Measurements of temperatures, pressures and solid withdrawals are performed at different axial positions in the reactor (3 positions for the riser and 5 for the outer annular region). Type B thermocouples with ceramic shields are used to measure temperatures in the riser. Type K thermocouples are used for all other temperature measurements. Concentrations of NOx, CO, CO_2, SO_2, O_2 and unburned hydrocarbons are obtained from an on-line gas analyzer.

Operating conditions and experimental results:

Spent foundry sand provided by a local foundry was used for the experimental work. Two types of spent foundry sand were used: core sand and mixed sand. Sand properties and resin characteristics are summarized in Table 1.

During a typical experimental run, the reactor is filled with clean sand and heated until steady state temperatures are reached. Spent foundry sand of known resin concentration is then fed to the reactor until steady state exit resin conversion is attained. The three experimental conditions studied in this work are presented in Table 2.

Table 1: properties of the spent foundry sand and resin

	mixed sand	core sand
Mean size, μm	180-190	180-190
Density, kg/m³	2600	2600
Phenolic resin % (wt)	2.2	3.2
C	<3.11	
H	<0.90	
N	<0.10	
S	<0.01	
O	2.05	
HHV, MJ/kg	0.6	0.9
Silica sand composition%(wt):		
SiO_2	96.44	96.44
TiO_2	0.00	0.00
Al_2O_3	0.15	0.15
CaO	0.055	0.055
MgO	0.036	0.036
Na_2O	0.015	0.015
K_2O	0.029	0.029
Fe_2O_3	0.207	0.207

Reactor Temperature:

Typical temperature profiles during the experiments are shown in Figure 3. At the start of the solids feeding period, the riser and annulus temperatures drop slightly. This profile is expected due to the fact that the heating value of the spent foundry sand is low. The power input due to the resin on 60kg/h of spent sand particles corresponds to 0.74MJ/h of, compared to 71.4 MJ/h for the combustion of 2.0 m^3N/h of natural gas.

Table 2: Operating conditions

	RUN 1	RUN 2	RUN 3
Spent foundry sand kg/h	20	40	60
Natural gas m^3N/h	2.1	2.1	2.1
Air, m^3N/h	27.5	27.5	27.5
Gs, kg/m^2s			
Total mass of solids treated during Run, kg	140	140	140

Resin removal:

The transient resin removal or conversion for the solid phase is plotted in Figure 4. It varies with waste feed rate or residence time in the combustion chamber. The solid circulation rate controls the number of cycles each sand particle passes through the riser. Under experimental conditions used in this work, the ratio G_sA_{riser}/W_{sand} is sufficiently high to achieve nearly complete solids backmixing in the reactor. Under these conditions, the influence of Gs cannot clearly be observed from these experimental data. Gas emission levels for the three runs are presented in Table 3.

Table 3: Stack emissions results

	RUN 1	RUN 2	RUN 3
Sand flowrate, kg/h	40	60	60
CO_2, %	6.7	7.8	7.1
CO, ppm	61	117	10–20
NO_x, ppm	48	29	20
O_2, %	7.6	5.4	6.7
Fines, %	<5	<5	<5
Combustion efficiency $CO_2/(CO_2+CO)$	0.9991	0.9985	0.9997

In general, emission levels are quite low. As in other fluidized bed systems, NOx formation is greatly dependent on the nitrogen concentration of the resin, hence the low NOx emission levels observed. Overall combustion efficiency was high. Fines generation was less than 5% of the total mass fed, which means that attrition is not important. In addition, the presence of an impact device and a disengagement zone at the riser exit helps the gas-solid separation and minimize particle transport. The change in particle size distribution was also measured since it is of critical importance to the founders in order to maintain a good quality of molds. Table 4 summarizes the change in those distributions before and after treatment. It is seen that the average particle size before the treatment is smaller than after the treatment, while the standard deviation is bigger. This is due to the treated sand having been striped of its fines by the fluidizing gas. It also proves that there was no significant breakup of sand grains. Molds fabricated with the treated sand showed excellent characteristics to the satisfaction of the foundry.

Table 4: Foundry sand size distribution summary

	Before reclamation	After reclamation
Median, µm	168	178
Mode at, µm	155	175
Mean, µm	181	189
Standard deviation, µm	113	81
Confidence, %	88.6	99.2

Comparison with other technologies:

The industrial potential of any new waste treatment process must be evaluated from both technical and economic aspects. The technical aspects include waste destruction and emission performances, while economical aspects are related to the treatment costs per ton of treated wastes. These costs include energy costs (in form of gas or electricity), investment costs, labor costs and maintenance costs. The energy cost is directly related to the performance of the technology and is referred to as the specific consumption of additional energy per kg of waste materials (kWh/kg). This economic factor was used to compare the present technology with other commercialized processes and the results are presented in Table 5. The value of 0.2 kWh/kg given for the ICFB is based on the maximum solids flowrate that could be fed to the pilot plant using the 20 kW natural burner. The limitation was in the solids feeding system itself that could not deliver higher flowrate to the unit, hence smaller specific consumption could be possible.

Table 5: Comparison of technologies

	Specific consumption (kWh/kg)
Rotary kiln	0.3
Fluidized beds	0.1–0.4 (with heat recovery)
Gas-contact	0.4–0.9 (no heat recovery)
Proposed ICFB	0.2 (no heat recovery)

Economical feasibility:

It was shown that the proposed process effectively removed the resin from the sand particles and that the treated sand can be recycled to make molds with retained properties. Table 6 below shows a payback time analysis for an industrial unit of 1 ton/h capacity. This assessment is based on a local foundry whose new sand cost is about 100$/ton and landfilling cost 25$/ton. The company uses 4000 tons of sand per year and dumps the same quantity. Costs associated with thermal reclamation unit come from different sources [Bex,1991; Bralower and Bardin, 1988; Lumsden 1993; Reier 1993]. For a typical foundry that generates 1000 to 6000 tons/year: energy costs (gas) is 14$/ton, operation costs (labor and maintenance) are 6$/ton, investment costs (within a 10 year period) are 22$/ton.

The payback period is less than 3 years for the considered foundry. This is mainly due to the high cost of new sand used (silica sand) and landfill costs. Payback time is expected to decrease as cost of landfilling continues to increase significantly in the future.

Table 6: Assessment of payback

	units	actual expense $/y	Reclamation at 100% reuse	Reclamation at 90% reuse
New sand	100$/ton	400 000	0	40 000
Landfill	25$/ton	100 000	0	10 000
Energy(gas)	10$/ton	-	40 000	40 000
Operation	6$/ton	-	24 000	24 000
Investment 880 000$	22$/ton	-	88 000	88 000
	Total Expenses	500 000	152 000	202 000
	Economy	-	348 000	298 000
	Payback	-	2.5 years	2.9 years

CONCLUSION

A novel thermal treatment technology for low heating value wastes has been tested at the pilot scale level. The first application dealt with reclamation of foundry sand. This waste is produced after several cycles of mold making and the resulting spent foundry sand particles is covered with an organic resin. Because of this resin, the waste is classified special waste and this leads to high landfilling costs, not taking into account the replacement cost of this sand for the foundries.

The internally circulating fluidized bed unit showed excellent performances for treating spent foundry sand. The high temperature contact between the solids and the flame region of a natural gas burner provided high combustion efficiency while maintained high overall energy efficiency, since only a small region of the reactor is kept at high temperature, that is the base of the riser. The remainder of the unit can be kept at lower temperature, which is not possible in conventional fluidized beds normally used for foundry sand thermal reclamation where the entire bed is maintained at the treatment temperature. The specific energy consumption is therefore very competitive for the ICFB and emission levels were low in CO and NOx. A brief economical assessment of ICFB for thermal reclamation of spent foundry sand showed relatively short payback times for a typical foundry.

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Figure 1 : Circulating fluidized bed reactor configurations

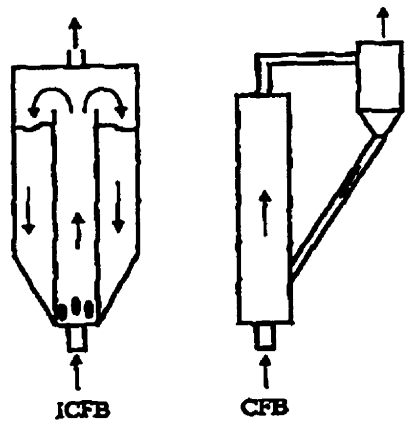
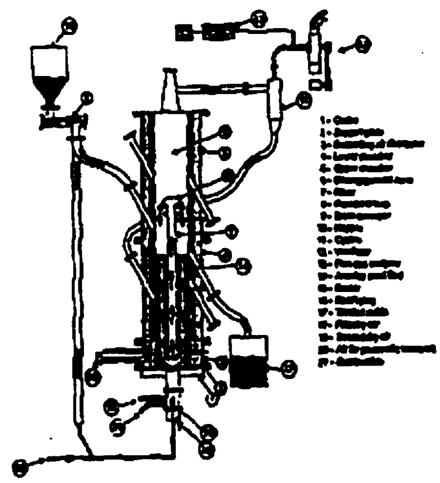


Figure 2 : The internally circulating fluidized bed pilot plant unit at the Greg Technology Research Group



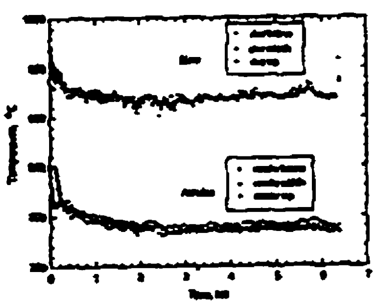


Figure 3 : Temperature profiles during the experimental runs

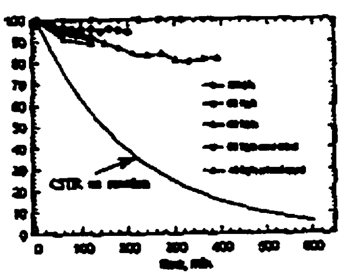


Figure 4 : Transient conversion of resin in the ICFB