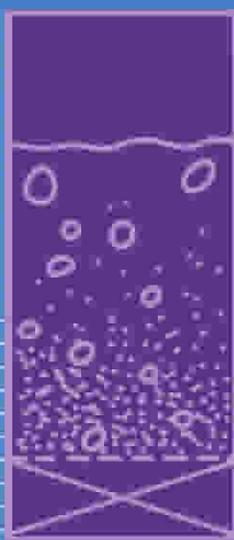
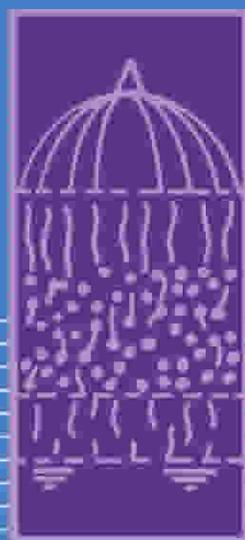


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In memory of my father
and to my mother

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Preface

The field of gas-liquid-solid fluidization, in the broad sense, encompasses any gas-liquid-solid system where solids are in a nonstationary state. Such systems have gained considerable importance as evidenced in their commercial or demonstrated applications in physical, chemical, petrochemical, electrochemical, and biochemical processing. Successful application of gas-liquid-solid fluidization systems lies in comprehensive understanding of the transport phenomena, including hydrodynamics, heat and mass transfer, and mixing. The transport behavior in these systems is, however, very complex. This book attempts to provide a unified treatment of the principles underlying transfer phenomena of various fluidization systems and to reflect their impact on reactor applications.

The book consists of three parts. Part I is an introduction which describes a unique method of classification of a wide range of gas-liquid-solid fluidization systems varying from fluidized beds, slurry bubble columns, and turbulent bed contactors, to three-phase transport. Numerous application examples using these systems are also given. Part II discusses transport fundamentals of gas-liquid-solid fluidization, described according to the classifications given in Part I. Here, mechanistic interpretations based on theoretical and semi-theoretical approaches to transport phenomena systems are described. Moreover, a comprehensive account of the empirical approaches in terms of correlations is given. In Part III, numerous industrial applications of three-phase fluidization systems including results obtained from large scale systems are presented in the context of the transport phenomena approaches given in Part II. Pertinent reactor engineering problems associated with these applications are also addressed. A classical approach to catalytic reactor analysis utilizing transport properties described in Part II is exemplified in Appendix A. Appendix B summarizes the SI units on which all the empirical correlations given in this book are based.

The book can be used as a graduate textbook in a three-phase fluidization engineering course or as a supplemental textbook to a general fluidization engineering course. It is desirable that students who use this book have already had some background in two-phase systems and are familiar with the classical material presented in, for

example, Wallis (1969) (One-Dimensional Two-Phase Flow, McGraw Hill) and Kunii and Levenspiel (1969) (Fluidization Engineering, John Wiley). The book can also be used as a supplemental textbook or reference for multiphase reaction engineering courses covering areas including biochemical engineering, environmental engineering, petroleum engineering and energy engineering. The book complements well books on general three-phase reactors by Shah (1979) (Gas-Liquid-Solid Reactor Design, McGraw Hill) and Ramachandran and Chaudhari (1983) (Three-Phase Catalytic Reactors, Gordon and Breach) which are more reaction engineering oriented. The book will also be useful to practicing engineers or industrial researchers who require readily available transport properties information for system design, operation or simulation.

Completion of this book would not have been timely without numerous very dedicated and capable members of my research group who have helped with the project over the years. I am especially grateful to Dr. K. Muroyama for providing an extensive literature survey, particularly on empirical correlations which are included in the tables and figures of Chapters 3, 4, 5, and 6. I owe much to Mr. D. C. Arters, Dr. Y.-M. Chen, Dr. K. Fujie, Mr. R. L. Gorowara, Mr. R.-H. Jean, Dr. K. Kitano, Mr. B. E. Kreischer, Dr. T. Miyahara, Mr. P. Murray, Mr. G. H. Song, Mr. W.-T. Tang, Dr. K. Tsuchiya, Professor K. D. Wisecarver, and Dr. Y.-S. Yang for providing literature material, and for making valuable original contributions which are directly incorporated in the text. Other research group members have read the entire text and made useful technical and editorial suggestions. My inquisitive students in the Chemical Engineering 815.15 course, entitled "Fluidization Engineering", have provided valuable feedback about some portions of the text.

I am deeply grateful to several colleagues who have reviewed a specific chapter in the application part in the area of their expertise and made significant comments: Mr. R. D. Buttker, Professor J. J. Chalmers, Dr. C. S. Chang, Mr. J. F. Mosby, Dr. L. E. Peck, Professor S. T. Yang and Ms. L.-P. Zhang. I express my special thanks to Professor M. A. Bergougnou, Professor N. Epstein, Professor J. P. Hartnett, Professor R. Leyva-Ramos and Mr. D. N. Smith, who have provided valuable suggestions and overviews on the text, and to many others who graciously responded to my requests for specific information for inclusion in this book. I want to especially acknowledge my colleagues in the Department of Chemical Engineering at The Ohio State University: Professor R. S. Brodkey, Professor T. L. Sweeney, and

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Finally, I am most indebted to my wife, Shu-Huan, for her support and enduring sacrifices throughout my academic career and to my children, Jonathan and Joline, for brightening the days.

L.-S. Fan
Columbus, Ohio

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Chapters 3, 4, 5, and 6 of this book were written, in part, based on a literature survey and pertinent comments provided by Dr. Katsuhiko Muroyama. He is duly credited as a coauthor of these chapters for his contribution. Dr. Muroyama is presently with the Department of Environmental Chemistry and Technology at Tottori University, Japan.

L.-S. Fan

Part I

Introduction

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Chapter 1

Classification and Significance

Gas-liquid-solid systems for physical, chemical, petrochemical, electrochemical, and biochemical operations can refer to a variety of operational schemes. Processing alternatives include the choice of continuous phase, relative flow directions, and continuous flow or batch. For example, both the liquid and solids can be either in continuous flow or batch while the gas is in continuous flow; the flow of gas and liquid can be cocurrent upward, cocurrent downward, countercurrent, or crosscurrent; the flow of solids can be upward or downward independent of or dependent on the gas or liquid flow; the gas may either be a continuous phase or discrete bubbles; the liquid may be a continuous phase, a film, or droplets; and the solids are a discrete phase in either a packed or suspended state (Ostergaard, 1968; Shah, 1979; Epstein, 1981; Shah et al., 1982; Muroyama and Fan, 1985; Smith et al., 1986; Fan et al., 1988). Because of this diversity, establishing a unified classification scheme for gas-liquid-solid systems is not an easy task. This chapter describes the classification of gas-liquid-solid systems, operating modes of fluidization, and applications of gas-liquid-solid fluidization and compares fluidization systems with fixed bed systems.

1.1 CLASSIFICATION

It is desirable to classify gas-liquid-solid systems through appropriate differences in hydrodynamic behavior. The hydrodynamic behavior is characterized by the type of operation, relative direction of flows, and continuity of the phases and is inherently related to the state of motion of the solid phase. Classification of three-phase systems can readily be extended from that of gas-liquid, gas-solid, and liquid-solid systems. It is, however, convenient and practical to classify them according to the state of particle motion in a

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manner similar to that for gas-solid or liquid-solid systems¹. The state of particle motion can be subdivided into three basic operating regimes: the fixed bed regime, the expanded bed regime, and the transport regime. The fixed bed regime exists when the drag force on the particles induced by the flow of a gas-liquid mixture is smaller than the effective weight of the particles in the system. When, with an increase in gas and/or liquid velocity, the drag force counterbalances the effective weight of the particles, the bed is in the state of minimum fluidization and marks the onset of the expanded bed regime. With a further increase in gas and/or liquid velocity beyond the minimum fluidization velocity, operation is in the expanded bed regime until the gas or liquid velocity² reaches the terminal velocity of the particle in the gas-liquid medium (U'_t). At a gas or liquid velocity above U'_t , operation is in the transport regime. An example of an operating regime map for an air-water-solid system with cocurrent upward flow of gas and liquid with liquid as the continuous phase is shown in Fig. 1.1. In this figure, the variations of the minimum fluidization velocity (U_{mf}) and U'_t with the particle terminal velocity in the liquid medium (U_t) and the gas velocity³ (U_g) are given. As shown, U_{mf} increases with increasing U_t but decreases with increasing gas velocity. For particles with a small U_t , U'_t is unaffected by gas velocity; but for particles with a large U_t , U'_t decreases with increasing gas velocity. Note that Fig. 1.1 was obtained for particles with uniform properties in the bed.

For gas-liquid upward flow with liquid as the continuous phase, fluidized bed and slurry bubble column systems are the two most extensively investigated three-phase systems involving a solid phase in suspension. A diverse range of operating conditions have been labeled slurry bubble column operation; indeed, the operating conditions for "slurry" systems are loosely defined. Typical operating ranges for three-phase fluidized beds and slurry bubble columns for air-water-solid systems are shown in Fig. 1.2 (Fan et al., 1987). Note that three-phase fluidized beds operate in the expanded bed regime (covering U_t from 3 to 50 cm/s in the figure) and slurry bubble columns may operate in both the expanded bed regime and the transport regime (covering U_t from 0.03 to 7 cm/s in the figure). Thus, overlap for U_t from 3 to 7 cm/s exists between fluidized bed

1: The classification described here is based on a vertical system.

2: It depends upon which phase is the continuous phase (see Section 2.3.5).

3: Throughout this book, the operating variables for the gas phase are expressed based on the prevailing operating conditions of the system unless otherwise indicated.

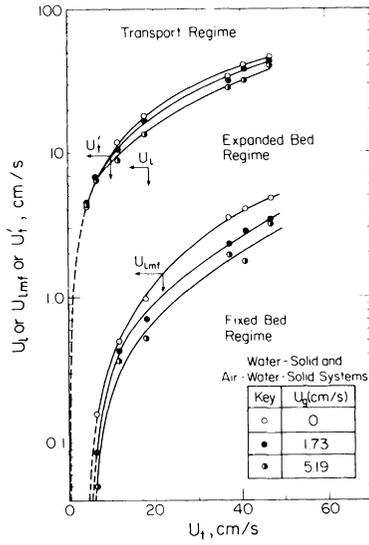


Figure 1.1 Operating regimes for cocurrent upward gas-liquid-solid systems with liquid as the continuous phase (from Fan et al., 1987).

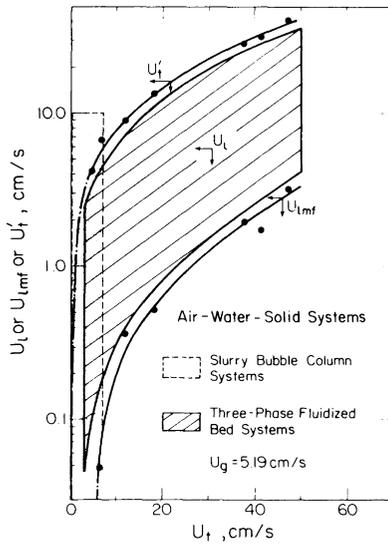


Figure 1.2 Common operating ranges for three-phase fluidized bed and slurry bubble column systems (from Fan et al., 1987).

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and slurry bubble column operations in the expanded bed regime. For convenience, the demarcation between the fluidized bed and slurry bubble column operated in the expanded bed regime is roughly set at 5 cm/s. In slurry bubble columns, the liquid velocity upper limit varies significantly with the application intended. In reactor applications, the upper limit for U_{ℓ} is usually set as high as 10 cm/s.

Generalization of the concepts leading to the operating regime map given in Fig. 1.1 is needed. Naturally, $U_{\ell\text{mf}}$ and U'_t vary with the relative flow direction and phase continuity of the systems. A regime map such as that given in Fig. 1.1 can provide a unified and rigorous classification scheme. Such a classification scheme permits appropriate bounds to be established for correlations to be employed without the undue extrapolation which has occurred with gas-liquid mass transfer correlations as noted by Kim and Kim (1986).

1.2 OPERATING MODES FOR FLUIDIZATION SYSTEMS

In a broad sense, gas-liquid-solid "fluidization" encompasses both the expanded bed and transport regimes with U_t ranging from 0.03 to 50 cm/s. Figure 1.3 gives the basic operational schemes, or modes, for each operating regime.

In the expanded bed regime with a continuous liquid phase flowing cocurrently upward with the gas, solids can be introduced either batchwise or continuously. For large or dense particles ($U_t > 5$ cm/s, see Section 1.1), the solids can be charged and discharged independent of the liquid flow (Mode E-I-a-1); in such a system, the particles are uniformly distributed, in a global sense, in the bed and the bed height is well defined within the system⁴. For small or light particles ($U_t < 5$ cm/s, see Section 1.1), the charge and discharge of solids usually depend on the liquid flow (Mode E-I-a-2); in such a system, the particle concentration is high at the bottom and progressively decreases axially upward and the bed surface is ill-defined. For both Modes E-I-a-1 and E-I-a-2, bed expansion can be supported by the liquid phase, the gas bubbles, or both. In the expanded bed regime with a continuous gas phase flowing cocurrently upward with the liquid (Mode E-I-b), the solids can be continuously charged and discharged independent of the gas flow; the particle bed is mainly supported by the flow of gas.

4: This behavior generally refers to systems without internals.

Expanded Bed Regime in Gas-Liquid-Solid Fluidization	Mode Designation	E-I-a-1	E-I-a-2	E-I-b	E-II-a-1	E-II-a-2	E-II-b	E-III-a	E-III-b	
	Schematic Diagram									
	Continuous Phase	Liquid			Gas	Liquid		Gas	Liquid	Gas
	Flow Direction	Cocurrent Up-Flow				Countercurrent Flow			Gas Up-Flow Liquid-Batch	
	References (Chapters)	1, 2, 3, 6, 7, 8, 10, 11, A	1, 4, 6, 7, 10, 11, A	1, 2, 11	1, 5, 6, 7, 8	5, 9, 11	1, 5, 7, 9, 11	1, 4, 7, 11	1, 5	
Transport Regime in Gas-Liquid-Solid Fluidization	Mode Designation	T-I-a-1	T-I-a-2	T-I-b	T-II-a	T-II-b	T-III-a		T-III-b	
	Schematic Diagram									
	Continuous Phase	Liquid			Gas	Liquid	Gas	Liquid		Gas
	Flow Direction	Cocurrent Up-Flow				Countercurrent Flow		Cocurrent Down-Flow		
	References (Chapters)	1, 6, 7, 8, 11	1, 4, 10, 11, A	1, 6	1, 9, 11	1, 9	1, 6, 11	1, 6, 9		

(S-->) Independent Charge of Solids from Fluid (-->S) Independent Discharge of Solids from Fluid (+S) Charge or Discharge of Solids with Fluid
A : Appendix A

Figure 1.3 Basic classification of gas-liquid-solid fluidization systems.

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In the expanded bed regime with the liquid phase flowing downward countercurrent to the gas phase, the direction of bed expansion depends on the density difference between the particle and the continuous phase; in these systems the solids are continuously charged and discharged independent of the gas and liquid flows. When the liquid density exceeds the solids density and the liquid phase is continuous, the particle bed expands downward supported by the liquid flow (Mode E-II-a-1); this mode of operation is referred to as that of an inverse fluidized bed. When the particle density exceeds the liquid density, the particle bed expands upward supported by gas bubbles (Mode E-II-a-2). When the gas is the continuous phase, the liquid density usually significantly exceeds the solid density and the liquid trickles down through an expanded bed of particles supported by the gas phase (Mode E-II-b). This mode of operation is typical of, but not limited to, that of the turbulent bed contactor (TBC); however, in a high solid density situation TBC operation could also be conducted under Mode E-II-a-2.

In the expanded bed regime with a batch liquid phase, the solids are charged and discharged batchwise. When the particle density exceeds the liquid density and the liquid is the continuous phase, the expanded bed of particles is supported by gas bubbles (Mode E-III-a). When gas is the continuous phase, a bed of particles expands upward supported by the gas phase (E-III-b); the liquid phase exists as a film or droplets. Depending on the gas velocity, the axial solids concentration distribution for Modes E-III-a and E-III-b operation may vary from an exponential decrease at low velocities to near uniformity at high gas velocities⁵.

In the transport regime, gas and/or liquid velocities exceed those in the expanded bed regime and continuous charging and discharging of solids usually accompanies the liquid flow. When the continuous liquid phase flows cocurrently upward with the gas and solids, particles with either large or small U_t can be transported mainly by the liquid phase (Mode T-I-a-1) and the bubbling gas phase can be used to establish a uniform particle distribution (Mode T-I-a-2). Mode T-I-a-2 represents an operational condition of the slurry bubble column where particles are usually charged or discharged continuously as slurries (a liquid-solid mixture). The system can also be operated with gas as the continuous phase (Mode T-I-b).

5: See footnote 4.

In the transport regime with a countercurrent flow situation, slurries flow downward relative to the upward flow of gas with either liquid as the continuous phase (Mode T-II-a) or gas as the continuous phase (Mode T-II-b). In cocurrent downward systems, the continuous phase can be liquid (Mode T-III-a) or gas (Mode T-III-b). Note that the flow with gas as the continuous phase shown in Fig. 1.3 is exemplified by an annular flow.

The presence of internals within a system, such as draft tubes, heat exchangers and baffles, or geometric irregularities such as a tapered geometry, greatly increases the complexity of the transport phenomena; however, the classification scheme given above is still valid for such systems. For the transport regime, the same mode designations also apply to situations where solids are initially present in the gas phase forming a gas-solid mixture; they then progressively transfer to the liquid phase as the mixture flows through the system. Although the general classifications are based on a vertical system, similar considerations for classification can also be made for a horizontal system involving gas-liquid-solid transport. Note that for all fluidization systems described above or generally in this book, solids suspension is considered solely induced by flows of gas and/or liquid phases without the aid of external means such as mechanical agitation (e.g., Nienow, 1984) or magnetic fields (e.g., Hu and Wu, 1987; Wu et al., 1988), unless otherwise noted. Information sources in reference to modes of operation and chapters are provided in Fig. 1.3.

1.3 FIXED BED SYSTEMS VERSUS FLUIDIZATION SYSTEMS

The modes of operation for three-phase fixed bed systems can be classified in a manner similar to those for fluidization systems. Fixed bed systems can be operated with either gas or liquid as the continuous phase in cocurrent upward flow, cocurrent downward flow, or countercurrent with downward liquid flow and upward gas flow.

In cocurrent downward flow systems, four types of flow can be distinguished depending on the relative magnitude of the gas-to-liquid flow rate: mist or spray flow, trickle flow, pulse or slug flow, and dispersed bubble flow. In both mist and trickle flow, the gas is the continuous phase and the liquid is a discrete phase. In slug flow, the liquid rich part of the slug, which is followed by the gas rich part, appears longer in length and greater in liquid content as the liquid velocity increases. As the liquid velocity increases at a moderate gas

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velocity the gas rich part of the slug disappears and dispersed bubble flow occurs where the liquid is the continuous phase and the gas is a discrete phase. Similar flow patterns can also be distinguished for cocurrent upward flow systems including mist or spray flow, pulse or slug flow, and dispersed bubble flow.

Fixed bed operation is of considerable industrial interest. Common fixed bed operations are in cocurrent downward and countercurrent trickle flows. Fixed beds in cocurrent downward trickle flow are generally used as reactors for such reactions as catalytic desulfurization and hydrocracking of petroleum residues and catalytic hydrogenation and oxidation of chemical feed stocks. Fixed beds in countercurrent trickle flow, on the other hand, are generally used as mass transfer devices with or without chemical reactions such as wastewater aeration and carbon dioxide absorption.

There are inherent differences in transport properties between fixed bed and fluidization systems. The major differences lie in that fluidization systems yield considerably higher macromixing and heat transfer in gas, liquid and solid phases than do fixed bed systems. For process applications, comparisons of these systems are complex and difficult. It requires consideration of not only the transport properties but also reaction kinetics for the process application intended. However, commonly acknowledged general advantages of fixed bed systems over fluidization systems, and vice versa, used as catalytic reactors (e.g., Germain et al., 1979; Shah 1979) are described in the following.

The advantages of fixed bed systems over fluidized bed systems are low macromixing, yielding small axial dispersion of phases and high reactant conversions for reaction kinetics favoring plug flow patterns; high controllability over product selectivity for complex reactions; low solids attrition and consumption, hence permitting precious metal catalysts to be used for the reaction. The major advantages of fluidization systems over fixed bed systems are high macromixing, yielding large axial dispersion of phases and high reactant conversions for reaction kinetics favoring completely mixed flow patterns; ability in achieving significant temperature uniformity without the aid of external means; ease in heat supply and removal and hence temperature controllability; low intraparticle diffusion resistance and external liquid-solid mass transfer resistance; ease in catalyst replacement and hence high controllability of catalyst activity and minimum flow maldistribution. Specific comparisons of advantages and disadvantages between fixed bed and fluidized bed systems

based on two individual process applications, namely, aerobic wastewater treatment and hydrotreating and conversion of residues are given in Chapters 8 and 10, respectively.

Extensive information regarding the transport properties and reaction engineering of gas-liquid-solid fixed bed systems is available in the literature: hydrodynamics and reaction engineering, especially relating to hydrogenation - e.g., Satterfield (1975), Hofmann (1978, 1986), L'Homme (1979), Shah (1979), Van Landeghem (1980), Ramachandran and Chaudhari (1983); reaction engineering relating to oxidation - e.g., Goto et al. (1977); design and modeling - e.g., Herskowitz and Smith (1983); mass transfer - e.g., Goto and Smith (1975); Charpentier (1976, 1981, 1986). Readers are referred to these references for details about three-phase fixed bed operation.

1.4 APPLICATIONS OF FLUIDIZATION SYSTEMS

Numerous applications of gas-liquid-solid fluidization systems exist which vary in size from bench scale to commercial scale and include all of the basic modes of operation given in Fig. 1.3. In these applications, the individual phases can be reactants, products, catalysts, or inert (Shah, 1979; L'Homme, 1979; Ramachandran and Chaudhari, 1983). For example, all three phases are either reactants or products as in coal liquefaction (Ashland Synthetic Fuels, Inc., 1984); the gas and liquid phases can be either reactants or products and the solid can be a catalyst as in hydrogenation of α -methyl styrene in a slurry of alumina-supported palladium catalyst (Satterfield et al., 1969); two phases can be reactants or products and the third phase can be inert as in ammonia scrubbing by a boric acid solution (Douglas, 1964); and the phases can be neither reactants nor products as in purely physical processes such as air humidification (Douglas, 1964). In the following, historical highlights of commercial or large scale operation of three-phase fluidization systems are described. In addition, numerous processing examples of these systems conducted on a relatively small scale are given to reflect the importance of three-phase fluidization applications.

1.4.1 Historical Highlights of Commercial/Large Scale Operation

The use of gas-liquid-solid fluidization systems in reactor applications can be traced back to the work of Bergius from 1912

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through 1926 on direct coal liquefaction or coal hydrogenation (Storch, 1945; Probst and Hicks, 1982). In his studies, pulverized coal-oil slurries were reacted with hydrogen at high temperature and pressure using slurry bubble column systems (Mode E-I-a-2). The study led to the development of a direct coal liquefaction process at a mini-commercial scale, i.e., Bergius-Pier process, for the production of motor fuels from lignite in Germany in 1927 with a capacity of 2,500 barrels per day (bbl/day) (Wenzel et al., 1986). It is interesting to note that the timing of commercialization of this process approximately parallels that of the first commercialization of gas-solid fluidization systems, i.e., Winkler process, for coal gasification developed also in Germany (Kunii and Levenspiel, 1969). In 1935, a mini-commercial process for direct coal liquefaction was also operated in England. During World War II, the commercial production using this process in Germany peaked at an average annual production of 4.2 million tons of mostly aviation gasoline (Donath, 1963). The direct production of liquid fuels from coal was phased out, however, following the end of the war.

Liquefaction of coal to produce liquid fuels could also be achieved indirectly from liquid phase reactions, involving hydrogen and synthesis gas derived from coal gasification, in the presence of a catalyst. The reactions known as Fischer-Tropsch (F-T) synthesis were discovered in 1926 (Frohning et al., 1977). F-T synthesis has been conducted in various types of reactors including a slurry bubble column reactor (Mode E-III-a) developed by Kolbel and Ackermann (1956) during the period 1938-1953 in Germany (see Section 11.9). The demonstration reactor developed by the companies Rheinpreussen and Koppers for the liquid phase F-T synthesis was operated from 1952-1953 at a daily production rate of 11.5 tons of liquid fuels (Kolbel and Ralek, 1980). Production was then discontinued due to the availability of cheap petroleum crudes and natural gas.

Though a bench scale study was first performed in the 1940's, it was not until 1987 that a slurry bubble column reactor (Mode T-I-a-2) was successfully proven at a mini-commercial scale in Canada for hydrotreating of tar sand and petroleum resid (see Chapter 10). The reactor is a part of the Canmet hydrocracking process which has a processing capacity of 5,000 barrels per day (bbl/day) (Lunin et al., 1985).

Slurry bubble column or transport reactors (Modes E-I-a-2, E-III-a, T-I-a-1, T-I-a-2, or T-III-a) have been successfully utilized as an alternative to gas-liquid-solid trickle bed reactors or gas-solid

fluidized bed reactors for catalytic production of organic chemicals or polyolefins since the 1950's. Slurry bubble column or transport reactors appeared in various designs and were operated over a wide range of flow conditions for catalytic reaction applications. The earlier production came mostly from Europe. Major reaction applications in commercial production (e.g., Germain et al., 1979) included hydrogenation of glucose to sorbitol, benzene to cyclohexane, benzoic acid to cyclohexanecarboxylic acid, butynediol to butenediol, adiponitrile to hexamethylene diamine, esters to fatty alcohols, 3,5 nitrotoluene to 3,5 toluene diamine, and aluminum and ethylene to Ziegler alcohol (ALFOL process), and ethylene polymerization (Solvay process using Ziegler-Natta catalysts). Clearly, hydrogenation reactions are typical of these applications.

The three-phase fluidized bed reactor (Mode E-I-a-1) was first used commercially in 1968 for hydrotreating petroleum resids (see Chapter 10). The process, known as the H-Oil process, was developed in the U.S. following research and development beginning in the late 1950's. Based on similar technology, the LC-Fining resid hydrocracking process was later developed in the U.S. Several commercial reactors based on either process are currently in operation in refineries in Kuwait and Mexico as well. The processing capacities in these refineries range from 18,500 to 60,000 bbl/day. The reactor is commonly referred to in industry as an "ebullated bed" where particles are in "ebullation" induced by gas-liquid phases. It is noted that the technical term "ebullated bed" or "ebullation" was first defined by P.W. Garbo in the patent of Johanson (1961) to describe a gas-liquid contacting process in contrast to the common industrial term "fluidized bed" where particles are in "fluidization" induced by the gas phase alone.

The energy crisis in 1973 brought renewed interest in the synthetic fuel area. Demonstration or semi-demonstration process units for direct coal liquefaction were developed in the U.S. These processes included the Exxon Donor Solvent (EDS) process and Solvent Refined Coal (SRC) process, and H-Coal process. The EDS and SRC processes use slurry bubble column (Mode E-I-a-2) reactors operated at a coal feed rate of 250 ton/day and 50 ton/day, respectively. The H-Coal process was developed based on the same technology as the H-Oil process using a fluidized bed reactor (Mode E-I-a-1) and was operated at a coal feed rate of 500 ton/day. All of these processes were, however, phased out in the early 1980's, again due to the low price of petroleum crudes.

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The energy crisis also led to innovative development in flue gas cleaning systems for coal combustion processes. A three-phase fluidized bed system (wet scrubbing system) known as the Turbulent Bed Contactor (TBC) (Mode E-II-b, mostly), first introduced in the late 1950's (Chem. Eng., 1959), was used for this purpose (see Chapter 9). In the mid-1970's, TBC scrubbers were operated at a commercial scale to remove sulfur dioxide and particulates from flue gas generated from coal combustion. Typically, each TBC had the operational capacity to treat 100 MW equivalent of flue gas containing sulfur dioxide and particulates. The flue gas scrubbing application of the TBC was terminated in the early 1980's, partly due to maldistribution and inconsistent durability problems associated with the low density particles used and the excessive pressure drop over a slurry spray tower (Mode T-II-b) (see Chapter 9). Since the late 1970's, commercial flue gas desulfurization has been conducted mainly using slurry spray towers.

Three-phase fluidization systems have made inroads into biotechnology covering biological wastewater treatment and fermentation. The fluidized bed bioreactor using immobilized living cells for wastewater treatment was conceived as early as the 1930's. Commercial application of three-phase fluidized bed bioreactors with immobilized living cells, however, was not demonstrated until the late 1970's and early 1980's, when the Biorex process bioreactors (Mode T-I-a-1⁶) with sizes ranging from 34 to 342 m³ were put in operation to treat night soil, tobacco mill and paper-board mill wastewater (Suzuki et al., 1981) (see Chapter 8). Development of immobilized living cell technology for fluidized bed or slurry bubble column bioreactor applications in fermentation significantly lags that in wastewater treatment. Although the initial success in cell immobilization for fermentation was demonstrated in the early 1960's (Linko and Linko, 1983), large scale three-phase fluidized bed or slurry bubble column fermentors were not demonstrated until the early 1980's, as exemplified by ethanol production in a 10,000 liter fermentor (Mode E-I-a-2) (Samejima et al., 1984) and penicillin production in a 200 liter fermentor (Mode E-III-a) (Endo et al., 1986), both in Japan (see Chapter 7). In the U.K. around the same time, a slurry bubble column bioreactor (Mode T-I-a-1⁶) of 1000 liter working volume was put in

6: The Biorex reactor is a draft-tube three-phase bioreactor. The mode designation given here is based on the behavior in the draft tube region of the bioreactor.

operation for the production of several kilograms per year monoclonal antibodies using suspended mammalian cells⁷ (Birch et al., 1985).

It is noted that in each of the large scale reactor operations described above, three-phase transport systems (operated in the transport regime) were naturally also in place on a large scale for reactant feeding, product discharging, internal/external fluid/solid recycling, etc.

1.4.2 Application Examples

Beyond those commercial or large scale applications given above, there are numerous examples of applications of three-phase fluidization systems demonstrated at relatively small scales. These examples are grouped according to physical, chemical (petrochemical or electrochemical) and biochemical applications as given in Tables 1.1 through 1.3, respectively. Tables 1.1 through 1.3 also show modes of operation for each application. It is seen that for all applications, modes in both the expanded bed regime and transport regime are used⁸. However, Modes E-I-a-1, E-I-a-2, E-II-b, E-III-a and T-I-a-2 are the most common modes for reactor application. Note that a given application may possibly be conducted in more than one mode of operation (e.g., hydrotreating of heavy petroleum residue using Modes E-I-a-1 and T-I-a-2). In some instances, a part of the system can be operated in a three-phase condition while the remaining part can be operated in a two-phase condition (e.g., urea granulation and production of polyethylene and polyolefins). In most aerobic fermentation systems (e.g., penicillin production), the liquid flow, a nutrient solution flow, is usually very small. Consequently, these systems are regarded as being batch with respect to the liquid flow. For some anaerobic fermentation systems with no inlet flow of gas, but with significant outlet flow of gas generated from metabolic reactions (e.g., ethanol production), the systems are regarded as being a special case of three-phase fluidization operation where the gas velocity in the feed is zero.

7: It is noted that for suspended cell systems which include those of microbial, plant and mammalian cells, only the plant and mammalian cells are treated fluid mechanically as being heterogeneous to the liquid phase as they are substantially larger than microbial cells. Hence, for suspended cell systems, only those involving plant and mammalian cells are considered in the context of three-phase operation.

8: For systems with non-uniform particle properties, the mode designation is based on the behavior of the average particle properties.

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Table 1.1 Examples of Physical Processing

Applications	Modes of Operation	Examples of References (Chapters or Sections)
Sand filter cleaning	E-I-a-1	Brownell (1951) (also see Sec. 11.4)
Crystallization	E-I-a-1 E-I-a-2	Thompson (1955) Okochi et al. (1978) (also see Sec. 11.5)
Air flotation	E-I-a-2	Fuerstenau (1980) Foot et al. (1986) (also see Sec. 11.6)
Urea granulation, simultaneous granulation and drying	E-I-b	Kono (1980) Morl and Kunne (1986) (also see Sec. 11.2)
Dust collection	E-I-b	Nienow and Killick (1983) (also see Sec. 9.2)
Air cooling, humidification or dehumidification	E-II-b	Douglas (1964) Barile and Meyer (1971) Barile et al. (1974) Adamiec et al. (1976)
Particulate collection	E-II-b	Mlodzinski and Warych (1975) Uysal (1978) (also see Sec. 9.2)
Lactose granulation	E-II-b	Waldie et al. (1987) (also see Sec. 11.2)
Drying of granular material	E-III-b	Palancz (1983)
Three-phase transport	T-I-a-1	Toda et al. (1982) Kim and Choi (1984) Kato et al. (1986) Hatate et al. (1986)
Lifting of solids in shaft, well, and tunnel drilling or lifting of coal with underground water; Pumping manganese nodules from below ocean surface (air-lift pump applications)	T-I-a-1	Weber and Dedegil (1976) Giot (1982)
Three-phase transport	T-I-b	Kato et al. (1986) Hatate et al. (1986)
Drying of calcium carbonate and polyvinylchloride particles	T-I-b	Martin and Saleh (1981)
Three-phase transport	T-III-a	Shah et al. (1983) Hatate et al. (1986)
Three-phase transport	T-III-b	Hatate et al. (1986)

Table 1.2 Examples of Chemical, Petrochemical, or Electrochemical Processing

Applications	Modes of Operation	Examples for References (Chapters or Sections)
Hydrotreating and conversion of heavy petroleum and synthetic crude (H-Oil, LC-Fining or ebullated bed reactor)	E-I-a-1 (with internal recycle tube)	Johnson et al. (1985) Beaton et al. (1986) (also see Chap. 10)
Coal liquefaction (H-Coal or ebullated bed reactor)	E-I-a-1 (with internal recycle tube)	Li and Liu (1981) Ashland Synthetic Fuels Inc. (1984)
Electrodes	E-I-a-1	Oloman and Watkinson (1975) Kusakabe et al. (1981) (also see Sec. 11.1)
Methanation	E-I-a-1	Blum and Toman (1977) (also see Sec. 11.8)
Production of zinc hydrosulfite	E-I-a-1	Sastri et al. (1983)
Hydrogenation of heptane	E-I-a-1	Ermakova et al. (1973)
Hydrogenation of 1-heptene	E-I-a-1	Gartsman et al. (1977)
Calcium bisulfite production	E-I-a-1 and E-I-b	Volpicelli and Massimilla (1965, 1970) (also see Sec. 11.3)
Methanol production	E-I-a-1 E-III-a T-1-a-2	Sherwin and Frank (1976) Studer et al. (1987) (also see Sec. 11.10)
Coal gasification	E-I-a-2	Cover et al. (1973) Kohl et al. (1978) Yosim and Barclay (1981) (also see Sec. 11.8)
Coal liquefaction (Exxon Donor Solvent Reactor)	E-I-a-2	Tarmy et al. (1984) Mueller (1985)
Oxydesulfurization of coal	E-I-a-2	Joshi et al. (1981)
Production of polyethylene and polyolefins	E-I-b	Jenkins et al. (1985, 1986) (also see Sec. 11.7)
Adiponitrile production	E-I-b	Kono (1980) (also see Sec. 11.2)
Flue gas desulfurization using limestone slurries	E-I-b	Beaver et al. (1982)
Flue gas desulfurization using limestone slurries	E-II-b	McMichael et al. (1976) Wang and Burbank (1982) (also see Chap. 9)
Absorption of bromine from a bromide-water mixture by sodium hydroxide and iron bromide solutions	E-II-b	Gel'perin et al. (1965, 1966)
Rectification of a binary ethyl alcohol-water mixture	E-III-b	Gel'perin et al. (1968)

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(Table 1.2 continued)

Applications	Modes of Operation	Examples for References (Chapters or Sections)
Ammonia, carbon dioxide and fluorine gases scrubbing	E-II-b	Douglas (1964) Wozniak and Ostergaard (1973) Adamiec et al. (1976)
Hydrogen sulfite desorption	E-III-b	Kudra et al. (1978)
Fischer-Tropsch synthesis	E-III-a	Schlesinger et al. (1951, 1954) Kolbel and Ackermann (1956) Kolbel and Ralek (1980) Deckwer et al. (1980, 1981, 1982) Satterfield and Huff (1981) (also see Sec 11.9)
Oxidation of sodium sulfite	E-III-a	Kato et al. (1973)
Oxidation of ethylene	E-III-a	Fujimoto et al. (1976)
Hydrogenation of cyclohexane	E-III-a	Sherwood and Farkas (1966)
Hydrogenation of acetylene	E-III-a	Heck and Smith (1970)
Oxidation of sulfur dioxide	E-III-a	Goto and Kojima (1985)
Hydrogenation of α -methylstyrene	E-III-a	Govindarao and Chidambaram (1986)
Methanation	E-III-a	Hammer (1979) (also see Sec. 11.8)
Absorption of carbon dioxide in sodium hydroxide solution	E-III-b	Furusaki et al. (1984)
Coal slurry and hydrogen transport in preheater of coal liquefaction	T-1-a-1	Shah (1981) Kornosky et al. (1982)
Leaching of copper ores	T-1-a-2	Reilly et al. (1982)
Hydrogenation of glucose	T-1-a-2	Germain et al. (1979) Brahme et al. (1984)
Hydrotreating of tar sand and petroleum resids	T-1-a-2	de'Bruijn et al. (1982) Lunin et al. (1985)
Flue gas desulfurization using magnesia slurries	T-1-b	Egan and Felker (1986)
Flue gas desulfurization using limestone slurries	T-II-a	Idemura et al. (1978) (also see Chap. 9)
Flue gas desulfurization using limestone slurries	T-II-b	EPRI (1987) (also see Chap. 9)
Catalyst slurry and hydrogen transport in ebullated bed downcomer for coal liquefaction and resid hydrotreating	T-III-a	Beaton et al. (1986) Ashland Synthetic Fuels, Inc. (1984) (also see Chap. 10)

Table 1.3 Examples of Biochemical Processing (also see Chapters 7 and 8)

Applications	Modes of Operation	Examples for References
Treatment of lactose wastewater	E-I-a-1* (tapered bio-reactor)	Scott and Hancher (1976)
Treatment of phenol wastewater	E-I-a-1* (tapered bio-reactor)	Holladay et al. (1978) Lee et al. (1979)
Treatment of thiocyanate and coal hydrocarbonization wastewater	E-I-a-1 (tapered bio-reactor)	Lee et al. (1979)
Treatment of slaughter house wastewater	E-I-a-1	Qian (1980)
Treatment of municipal sewage wastewater	E-I-a-1	Shen and Wang (1980)
Treatment of phenol wastewater	E-I-a-1	Hirata et al. (1982) Hosaka et al. (1985)
Treatment of glucose-BOD wastewater	E-I-a-1	Takase et al. (1983)
Treatment of gasification wastewater	E-I-a-1	Donaldson et al. (1984)
Treatment of synthetic milk wastewater sewage	E-I-a-1* (tapered and non-tapered bio-reactor)	Forster et al. (1986)
Treatment of s-triazine wastewater	E-I-a-1* (tapered bioreactor)	Hogrefe et al. (1986)
Treatment of isobutanol or acetic acid wastewater	E-I-a-2	Chatib et al. (1981) Grasmick et al. (1981)
Ethanol production by yeast cells	E-I-a-2	Samejima et al. (1984)
Conversion of sucrose to glucose by plant cells <i>D. carota</i>	E-I-a-2	Prenosil and Pedersen (1985)
Bioprocessing of coal for liquefaction and removal of sulfur, nitrogen and trace metals	E-I-a-2 E-III-a	Wilson et al. (1986) Narayan (1987)
Simultaneous selective flocculation and microbial desulfurization of high sulfur coal	E-I-a-2	Fan and Attia (1988)
Treatment of phenol wastewater	E-II-a-1	Edwards (1981)
Treatment of oil refinery or kitchen wastewater	E-II-a-1	Hasegawa (1982) Shimodaira and Yushina (1983)
Ferrous iron oxidation by <i>Thiobacillus ferrooxidans</i>	E-II-a-1	Karamanev and Nikolov (1988)
Ethanol production by <i>Saccharomyces cerevisiae</i>	E-II-b	Moebus and Teuber (1982) Bauer (1986)
Bioleaching of metals from ores	E-III-a	Karaivko et al. (1977)
L-isoleucine production by <i>Serratia marcescens</i>	E-III-a	Wada et al. (1980)