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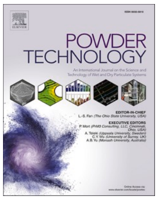

Hundred years of fluidization for the conversion of solid fuels

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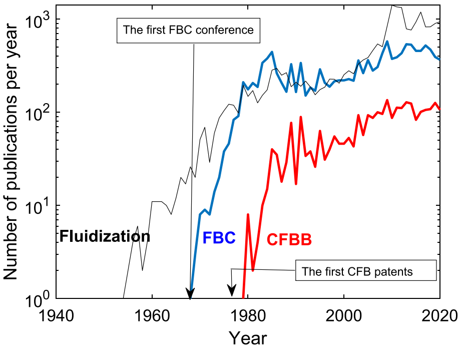
HIGHLIGHTS

- Fluidization was proposed in a patent for gasification of coal 100 years ago.
- It took about 50 years after the patent before it was introduced for combustion.
- Circulating fluidized bed proved the most suitable device for combustion of coal.
- Non-circulating beds are frequently used for combustion of biomass.

GRAPHICAL ABSTRACT



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ABSTRACT

This is a summary of the development of conversion of solid fuels in fluidized bed during the hundred years that follow the first patent of Winkler in September 1922. The Winkler gasifiers and their followers are described first. Other fuel converters, such as boilers, appeared only in the 1960–70s and became of interest because of their expected environmental advantages. Initially, bubbling bed boilers were introduced, followed by circulating fluidized bed (CFB) boilers in the beginning of the 1980s. Now, CFB is the dominant technology. The entire development has not been conditioned by technological breakthroughs, but rather by the surrounding conditions: industrial demand, wars, environmental effects, availability and price of fuels. The recent development of the presently rather mature technology depends very much on the necessity to limit greenhouse gas accumulation in the atmosphere. Although fluidized bed technology offers solutions to reduce CO_2 emissions, so far, no decisive line of application has been established for CO_2 reduction, except for the use of biomass and waste.

1. Introduction

Hundred years ago, on September 28th’ 1922, Fritz Winkler applied for a patent on a process for production of water gas [1]. A few years later, 1926, when the patent was granted and the design was ready, the first fluidized bed (FB) gasifier was put into operation. In the conical bottom of the gasifier a layer of coke (Grude) was heated by combustion with a stream of air that made the fuel powder show a vivid dancing movement that later was called fluidization. At a temperature of about 1100 °C the air was stopped, and steam was introduced with a sufficient strength to swirl the fuel bed, while producing the desired mixture of hydrogen and carbon monoxide gas without the use of further oxygen. Then, this procedure was repeated. This intermittent supply disturbed the performance of the bed, and in later applications it was replaced by

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an even addition of a mixture of air (or oxygen) and steam.

The patent referred to manifests the first step of industrial utilization of FB gasifiers to be followed by many more, and later it influenced the introduction of FB combustors. The development of fluidization for the conversion of solid fuels to be outlined below, was, like all technical applications, influenced by the surrounding conditions like war, supply of (alternative) fuels, environmental impact both locally and globally, fluctuations in fuel prices (for instance, cheap oil during 1950s and 60s followed by the sudden increments in price causing the “oil crises” in 1973 and 79), the economic situation in various countries etc.

Because of its significance as a source of inspiration, the Winkler gasifiers will be described first in some detail. Then other gasifiers and the emerging FB combustors and their relationships will be treated. Fluidization became applied in many areas. To focus the presentation, the following survey predominantly deals with the conversion of solid fuels. Surveys related to application of FB in the petroleum industry, another very significant application of FB, are found in [2,3], for example. A summary in the form of a timeline concludes the presentation.

2. Gasifiers

2.1. *Early Winkler gasifiers*

There were about 40 Winkler gasification units, operated between 1926 and 1975, mostly erected in the vicinity of lignite mines in Germany. Lignite (Braunkohle) and a coke residue of tar production from lignite, called Grude, were initially the principal fuels. There was a continuous development, and the various gasification units built differ slightly from each other, as can be seen from a few designs shown in Fig. 1.

Fig. 1a,b,c illustrates Winkler gas generators of different designs. Fig. 1d is a later version, the so called High-Temperature Winkler (HTW) gasifier, developed during the years 1976 to 1997. The earlier designs, Fig. 1a and b, are with and without a bottom gas-distributor plate. The second one has a simple cyclone for fly-ash recirculation [4]. After a few years of experience, it was considered that the bulb, forming the freeboard, complicated the operation and in later versions the walls were straight, although narrower in the bottom (Fig. 1c). Characteristic for all Winkler gasifiers is the split of air (or O_2) plus water vapour into a bottom supply and a secondary supply of these gases to the freeboard a few metres above the bed.

Sella [5] describes the discovery made by Winkler like this “It was while passing blowing air through a column containing coal powder at red heat that Winkler made a startling observation. As the flow of air was increased, a critical point would be reached where the coal particles were suddenly suspended by the flow and the whole mass appeared to boil. The particles began ‘a lively dancing movement’ in the flow and the whole mass became like a fluid. Bubbles could be seen rising through the mixture while denser objects dropped in at the top, sank, just as they would in a liquid of lower density.” In fact, the idea solved a problem with the lignite or Grude fuels used. The fuels tended to produce fines, which disturbed the operation of the fixed bed gasifiers usually applied for gas production. The remedy was to make briquettes for feeding to the fixed bed gasifiers, an expensive procedure. With the invention, a finely dispersed form was more reasonable for the conversion of the lignite fuels available in Germany.

The technical details of the Winkler gasifiers can be found in intelligence reports prepared after the end of the second World War (WW II) by the British and American authorities [6,7]. The data presented in the following regarding the Winkler gasifiers are taken from these reports. It should be realized that the various gasifiers differ a little from each other in design and operation and the data presented are typical rather than generally valid ones.

The gas qualities produced were an air-blown “producer gas”, a low-quality gas that could be supplied to boilers for combustion and an oxygen-blown “water gas” to be used for the manufacture of hydrogen, methanol, Fischer-Tropsch synthesis gas, or ammonia synthesis gas, in all cases based on a cheap fuel. Tables 1 and 2 show summaries of data [6].

The bulk of the fuel with a size of 0.2 to 4 mm was fed to the bottom part of the gasifier to give a bubbling bed height of 1 to 1.5 m of fuel, maintained at as high a temperature as the softening point of the ash permitted, around 950 °C. 10 to 33% of the $O_2 + H_2O$ added was introduced as secondary gas a few metres above the bed.

The fluidization velocity (u m/s) is not mentioned in the literature, but it can be estimated from Tables 1 and 2 for a large Leuna gasifier, accounting for the moisture content in the raw gas m, the staged

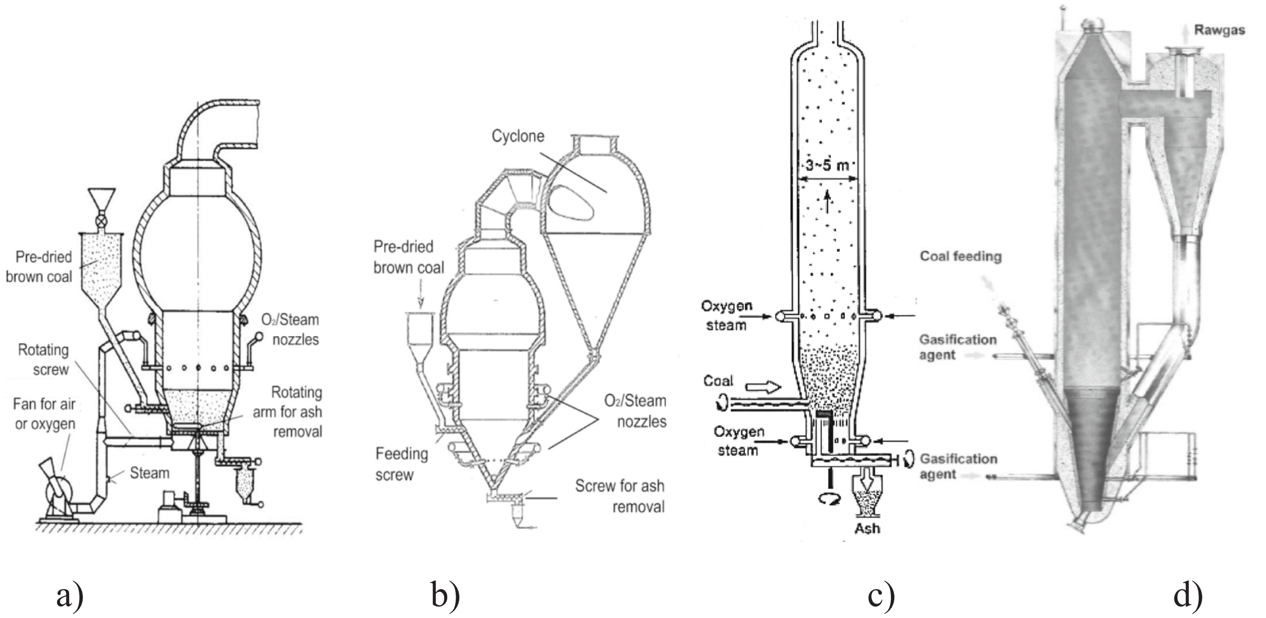


Fig. 1. a) An early version from the 1930s with an arm to remove the ashes; b) A device from the 1940s with a conical bottom and a cyclone for ash recirculation; c) A more modern gasifier after removing of the bulb; d) A High-Temperature Winkler gasifier.

Table 1
Design data of Winkler gasifiers making water gas, selected from [6].

Plant location	Leuna		Böhlen	Zeitz
Output of raw water gas (G), m_N^3/h	(Small)	(Large)		
Maximum	40,000	80,000	25,000	22,000
Normal	30,000	60,000	20,000	18,000
Minimum	–	–	12,000	9000
Fuel	Grude		Grude	Grude
Reactor shape	Some reactors are bulbous at top, some are straight sided. 1 small and 1 large reactor are grate-less. The remaining have a stationary grate.		Straight sided. Stationary grate.	Straight sided. Stationary grate.
Inner diameter of fuel bed (d), m	3.9	5.5	4.5	4 to 4.5
Height of fuel bed, m	1	1	1.5	1.5
Overall height of reactor (h), m	ng.	ng.	20	20

ng. —not given. “Grate” is nowadays called “distributor plate”.

Table 2
Operation data of a Winkler gasifier at Leuna, selected from [6].

Product	Water gas		Power gas	
Fuel	Grude	Brown coal	Grude	Brown coal
Raw gas output, m_N^3/h	55,000	27,000, 42,000	50,000	50,000
Proximate analysis, % as received:				
Combustibles	72	81	About the same as for water gas	About the same as for water gas
Ashes	26	13		
Moisture	2	6		
Elemental analysis, % dry-ash-free:				
Carbon	92.5	70.9	About the same as for water gas	About the same as for water gas
Hydrogen	2.7	5.4		
Oxygen	3.0	19.7		
Nitrogen	ng.	0.1		
Sulphur	1.8	3.8		
Analysis, raw, dry gas[*], vol%				(averages)
CO	37.5	38	32.7	22
H_2	39.5	40	7.6	12
CH_4	1.5	2	0.5	0.7
CO_2	20	19	4.3	8
N_2	0.5	1	54.6	55
H_2S	1.0	ng.	0.3	0.8
H_2 + CO, % of dry raw gas	77.5	78	41.3	34
Average fuel size, mm	3	3	ng	ng
Oxygen in O_2/H_2O, %	40–50	40	ng	ng
Secondary O_2/H_2O, G_{sec} %	33	33	ng	ng
Fuel bed temp. (T_{bed}) °C	850 to 900	800 to 950	ng	950
Gas exit temp. (T_{exit}) °C	900 to 950	950 to 1000	ng	1000
Dust in exit gas, kg/m_N^3	0.10 to 0.20	0.11 to 0.17	0.065	ng
Carbon in dust, %	50 to 55	29 to 35	ng	33–55
Carbon utilization, %	88	83	ng	83

[*] H_2O % on total gas can be estimated to 19% of wet raw gas [8]. ng—not given.

addition of oxygen and steam G_{sec}, and the temperature of the bed T_{bed}.

$$u = \frac{G^*(1+m)(100-G_{sec})/100}{3600^*\pi d^2/4}{}^*\frac{T_{bed}+273}{300} = \frac{55000^*1.19^*0.66^*1150}{3600^*25^*300} = 1.8\ m/s \tag{1}$$

The reactor volume above the bed is said to be 15 times of that of the bed, allowing a gas residence time of about 7 s. Assuming a freeboard height of a little above 20 m [7] and accounting for the secondary oxygen, a gas velocity in the freeboard of about 2.8 m/s results. This agrees rather well with Odell [7] who was informed that the gas velocity above the bed in the gas generators at Leuna was typically about 3 m/s. From the dry gas production at Zeitz [7] the velocities 1.8 to 4 m/s and at Leuna U = 0.8 to 2.4 m/s @ 1200 K can be estimated, but the values are from various sources and combined they become rather uncertain.

There was no other bed material than fuel, but the fuel contained 15–20% of ash on a dry basis, and the ash served as a bed material together with the combustibles. The bottom ash extracted was 10 to 20% of the total ash supplied and contained 30 to 55% of carbon. The fly ash of size 0.1 to 0.5 mm was collected and burnt in a neighbouring boiler. At Leuna a solids quantity of 60 g/m_N^3 in the exit from the gasifier has been mentioned. A particle separator, returning dust from the exit gas to the gasifier, is drawn on several sketches, but it was quite inefficient, and recirculation was not used in later designs, maybe because the fly ashes were utilized for combustion in nearby boilers. The bottom distributor caused many problems, and several versions are seen; two on Fig. 1 (a and c) including a stirring device activated once a minute (Fig. 1a), or later, a conical bottom for extraction of ashes and supply of steam and oxygen from the sides (Fig. 1b). The addition of oxygen and steam above the bed is seen in all cases of Fig. 1. This is to increase the gas temperature somewhat to enhance the rate of reactions and to utilize the space above the bed for conversion of the fuel.

2.2. High-temperature Winkler gasifiers

The original Winkler gasifiers without recycling of ashes were quite inefficient, reaching a fuel utilization (in the gasifier) of a little above 80%, but this was not of extreme importance under the given circumstances; the fuel was relatively cheap, and the ashes with their high content of char were used as a fuel. There was no reason to improve the situation during and just after the WW II. However, the gasifiers were taken out of operation gradually, and the year 1975 the last Winkler gasifier was stopped. Only a few years later a new development took place in Germany with the purpose of improving the performance of the gasifier. The new design was called High-Temperature Winkler (HTW) gasifier [9,10].

The HTW gasifier was introduced with the intention of improving the previous Winkler gasifiers by extending the range of application and the efficiency. The laws of nature (the melting properties of the ash) stopped the attempts to increase the temperature, and the temperature levels were about the same as in the previous designs, but the denomination “High Temperature” was retained in the name. One essential change was a better cyclone for dust recirculation, allowing a considerable improvement of the gasification efficiency from a little above 80% to above 90%. Another significant change was a higher pressure, increasing the throughput, and aiming at applications in the chemical industry (10 bar) and power production (25 bar). The pressure should also be beneficial for the rate of gasification.

In 1978 a 1.8 t/h pilot plant was built and operated till 1985, followed in 1986 by a demonstration plant in Berrenrath operating at 10 bar with a capacity of 37,000 m_N^3/h, producing gas for a methanol synthesis plant [11]. 1989 to 1992 investigations were carried out in a pilot plant in Wesseling, operating at pressures up to 25 bar, and so increasing the throughput from 5 t/m^2,h at 10 bar to 22 t/m^2,h at 25 bar to be applied in power production [12]. A typical dry gas composition was CO—45%; H_2—34%; CH_4—4%; CO_2–17%.

2.3. Other autothermal FB gasifiers

The Gas Technology Institute (USA) started 1974 to design a bubbling bed gasifier, called “U-gas process”, that after several years of development could operate at pressures ranging from 3 to 55 bar with steam and air or oxygen at temperatures from 840 to 1100 °C [13] with a variety of fuels. If the temperature was above the melting temperature of the ash, molten ash particles are formed. These ash particles tend to contain little combustible material and could be removed through an exit in the cone-formed bottom through a venturi separator, predominantly separating the ash particles. In this way the gasifier reduced the burnout problem that remained in the Winkler gasifiers when an efficient cyclone was employed—the ash must then be removed through the bottom, and if it is well mixed with the fuel, the loss of unburned will be considerable [14,15]. The coal utilization of the U-gas process has been reported to be 95% [13,15].

Many gasifiers were proposed throughout the world. For instance, a gasifier of “Winkler-type” was developed by Valmet (and previous companies incorporated in Valmet). The Valmet gasifiers [16] are very simple air-blown gasifiers, operating with waste wood or sorted municipal solid waste (MSW). An example is the 2 × 80 MW_{fuel} gasifier erected the year 2013 in Lahti, Finland, based on previous experience from from the Swedish boiler manufacturer Götaverken in the early 1980s, making FB gasifiers to serve lime kilns in pulp and paper companies (e.g. Värö 1987). The Finish company Tampella also built gasifiers in 1996, both being companies, which together with Metso, became incorporated in Valmet.

MSW contains a certain amount of coarse irregular objects. These objects must be sorted out and shredded to obtain a suitable size, while most metals and glass are removed before conversion in an FB. The form of the Valmet reactor reminds of the HTW gasifier on Fig. 1d, (circular cross section, 8 m diameter, 36 m height), but there is no secondary oxygen, and no steam is added. The air burns the volatiles to produce the desired temperature. Also, the small amount of char in MSW is burned according to.

$$(1-f)C + O_2 = (1-f)CO_2 + 2fCO; \tag{R1}$$

$$f = \frac{CO/CO_2}{2 + CO/CO_2} \tag{R2}$$

where f is a mechanism factor (if only CO_2 is produced f = 0; if only CO, f = 1). Normally, some CO is produced together with CO_2. However, this gasifier is mostly a devolatilizer, since at least 80% of the organic part of the fuel consists of volatiles, and only a minor part of the char is converted to CO as indicated from the reactions. The scheme in Fig. 2 explains the components of the gasifier.

Gas treatment to remove tar is needed in all biomass gasifiers, but here a special situation prevails due to the conditions of the European Union (EU) related to waste conversion [17], encouraging a certain efficiency of conversion even in the case of waste. In this case the product gas is cooled after the gasifier to 400–500 °C slightly above the condensation temperature of most volatiles but below the evaporation temperature of certain potentially deleterious alkali compounds, which are removed by high-temperature filtering. For these reasons the gas entering the combustor is relatively seen harmless and no longer a waste, since it has passed the legal criterion of “End-of-Waste”, established by the EU, and the downstream boiler is a flame-combustion boiler with conventional steam data (higher than in waste boilers). From a technical point of view, it is an interesting arrangement, but economically it may not be so attractive.

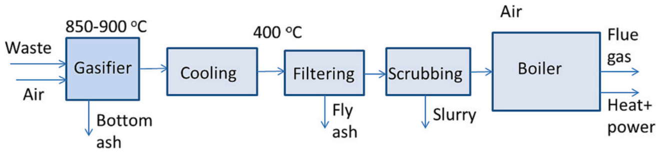


Fig. 2. The components of the conversion system in the Lahti plant.

3. FB combustors

Judging from the number of patents applied for, Standard Oil (USA) was one of the most active organizations developing FB already before the WWII. According to Squires [18], Odell, quoted above for his investigation on the Winkler gasifiers, was the first to use the word “fluidization” in written form (in a patent application). Subsequently, it became only gradually in use before it was generally accepted. In fact, not even in 1940 it was found in patents; Lewis and Gilliland [19] when describing a proposed FB catalyst cracking system used expressions like “a kind of quicksand” and compared the particle bed of their system with a boiling liquid.

In the development of Standard Oil there could have been influences from Germany. It is likely that the American oil companies knew about the activities of Winkler already in the 1930s when, according to Squires [18], they had some interaction with IG Farben, the German company behind the development of the Winkler gasifiers. On the other hand, Squires [18] quotes a statement from two of the principal developers at Standard Oil, the MIT professors Lewis and Gilliland: “Neither Dr Lewis nor I have ever seen a Winkler gas producer, and it was not the basis for our discussion”. Despite this statement, these inventors refer to Winkler in their patent applications. Some knowledge on the European activities must have been present. The activity of Standard Oil and other related companies was directed towards refining of oil, and although there were some early boiler patents [20], nothing of practical interest came out related to FB boilers.

3.1. Combustor conditions

The heat developed from F kg/s fuel supplied to an FB reactor, ψFH_{LHV} (fraction of fuel converted ψ and the lower heating value of the fuel H_{LHV}), is spent on heating of the gases from the initial temperature T_o to the bed temperature T_{bed} and on heat extraction by heat-transfer surfaces q,

$$Fgc_{pm}(T_{bed} - T_o) + q = \psi FH_{LHV} \tag{2}$$

which gives the bed temperature

$$T_{bed} = (\psi FH_{LHV} - q)/\left(Fgc_{pm}\right) - T_o \tag{3}$$

The quantity of flue gas evolved per kg fuel (g kg/kg) is calculated from the stoichiometry.

Gasifiers and combustors are designed and operated in different ways. For ψ = 1 (all fuel is burned) and q = 0 the bed is adiabatic, and the bed temperature is theoretically equal to the adiabatic one. That is above two thousand degrees, higher than the possible operation of an FB. Therefore, based on Eq. (3) there are two approaches of design to moderate the bed temperature: in a gasifier q = 0 (no heat is extracted) and ψ is in the order of 0.2 to 0.5, depending on the heat losses, to yield a temperature of 850 to 1000 °C, and for a combustor ψ = 1, which means that heat transfer surfaces are inserted in contact with the bed for heat extraction q to give the intended bed temperature.

In FB combustion devices the fluidization velocity (u) times the furnace cross-section area (A) is coupled to the combustion of F kg/s of fuel yielding a gas flow through the furnace equal to the gas released during the combustion

$$Fg = uA\rho \tag{4}$$

$$\rho = \rho_o PT_o/P_o T \tag{5}$$

where ρ_o is the gas density at pressure $P_o = 1$ bar and temperature $T_o = 273$ K.

The surface power Q ($W_{fuel}/m^2_{cross\ section}$) is the rate of fuel converted per unit furnace cross-section area, defined as

$$Q = FH_{LHV}/A$$

With Eq. (4) this is

$$Q = u\rho H_{LHV}/g \tag{6}$$

At a given fuel, temperature, pressure, and excess air ratio (included in g), the only parameter that can be varied is the fluidization velocity. A higher surface power means a smaller (cheaper) boiler, and therefore the designers of boilers try to employ high fluidization velocity, fighting the resulting carryover of particles.

Clearly, an increase of pressure also reduces the size of the furnace (but not necessarily the cost). Hidden in the specific gas quantity g is the fact that an increase of oxygen concentration at constant fluidization velocity in oxycombustion also leads to a smaller plant, because less nitrogen is substituted by diluent flue gas at higher oxygen concentration (in the case of flue-gas recirculation in oxy-fuel combustion). Eq. (4) shows that a smaller g leads to a smaller A, and hence, to a smaller furnace at constant fuel power and fluidization velocity.

3.2. *Bubbling FB combustors (BFB)*

The first activity related to combustion in FB, known to the author, was the effort to develop a combustor at the Moscow Energy Institute in the beginning of the 1950's [21]. Many German lignite mines were in the Soviet occupation zone in Germany where most Winkler gasifiers operated. It is likely that the Soviet intelligence service acted in the same way as their Western colleagues, extracting as much information as possible of German technology, among other topics that of the Winkler gasifier. Based on this assumption, the early development steps become understandable: the Soviet investigators treated the FB combustor as a gasifier operated at stoichiometric or over-stoichiometric oxygen supply ($\psi = 1$ but $q = 0$). The result was several years of struggle to handle the bed temperature that tended to become too high, causing sintering of the bed material. Before the end of their 10 years' effort, they had realized that the solution was to remove heat from the bed by cooling tubes ($q > 0$), thus attaining a suitable bed temperature. However, at that time the work finished because it was decided by the political authorities that the intended application, plants for district heating in the cities, should be provided by natural gas for environmental reasons. Further application of FBC in the Soviet Union was directed towards coal mines to burn mining wastes and for treatment of metals. This became the limited field of practical interest, despite the important research on fluidization going on in the country. It should be emphasized that the Soviet scientists were as aware of the possibilities of FBC as their colleagues in the West, which is illustrated in the *Foreword* to a study on heat and mass transfer by Baskakov et al. [22].

A very vivid activity in building FBC was initiated in the UK already in the 1960's and an impressive number of small boilers were introduced [23,24]. Encouraged by the possibility of burning difficult coals with sometimes high content of sulphur that could be removed by adding limestone to the bed, also USA got involved in the early development. An example of an early FBC in the UK, was the Renfrew 12 MW_{th} boiler (Fig. 3a) where the bed was inserted in an existing boiler. Typical for the units developed in the UK is their small size and strikingly low freeboard. It was common at the time to assume that the combustion takes place in the bed and not in the freeboard, which was quite reasonable because the main fuel was coal. The subsequent boilers did have a taller freeboard, but otherwise the design principles were the same, following the tradition of boiler design. The beds were cooled by immersed heat transfer tubes, and it was commonly emphasized that the heat transfer in a fluidized bed was very good.

In Germany a corresponding rebuilt FB boiler, Flingern, was put into operation in 1979 [25,26]. It was successful, and in Germany the interest for FBC increased. Like elsewhere, many different proposals were turned into boilers. Fig. 3b is just one example that is of particular interest because it is a bubbling FB boiler with recirculation of particles to the bed by means of a cyclone. In fact, earlier versions of this boiler type had cyclones to capture the fly ash, but it was, like in most designs at this time, not recirculated to the bed [26].

Likewise, FBC became of considerable interest in China where a 14 t/h (steam) boiler was built at the Tsinghua University already in 1969 [27]. FBC was suitable for available Chinese coals of low quality, and already in 1980 there were 2000 small bubbling FBC boilers in operation.

Initially, many designs of small bubbling-bed industrial boilers were proposed. Fig. 3 shows a selection of boilers built in the first wave of interest during the two decades 1970–1980 when bubbling beds dominated.

A great deal of investigations was carried out in laboratories to understand the fluid dynamics of fluidization, combustion, heat transfer, and environmental performance of bubbling beds. Small test units were operated, and the knowledge increased. However, the most essential obstacle to the operation of the proposed designs was only discovered when it was too late: after a few thousand hours of operation of a boiler, it became clear that, for the same reason that heat transfer to the immersed tubes was good, so was the erosion on the tubes. Fig. 4 shows an example of erosion on the in-bed tube bundles of the boiler in Fig. 3d.

It is clear from Fig. 4 that the erosion was most severe on the external tubes at the bottom and front where the tubes were most exposed to the

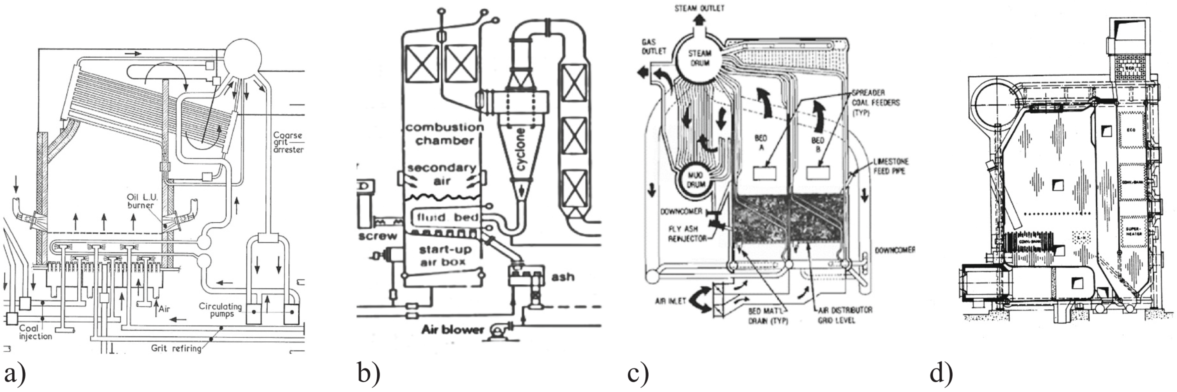


Fig. 3. Examples of early BFB boilers. a) The 12 MW_{th} boiler at Renfrew, retrofitted from an older boiler by Babcock Power Ltd. 1975. By 1979, 5000 h of operational experience had been acquired [28]. b) An early BFB plant from Germany [29]. c) The 32 MW_{th} steam boiler at Georgetown University, USA, 1980 [30]. d) The 16 MW_{th} boiler at Chalmers University, Sweden put into operation 1982, built by Generator AB (now Valmet) [31].

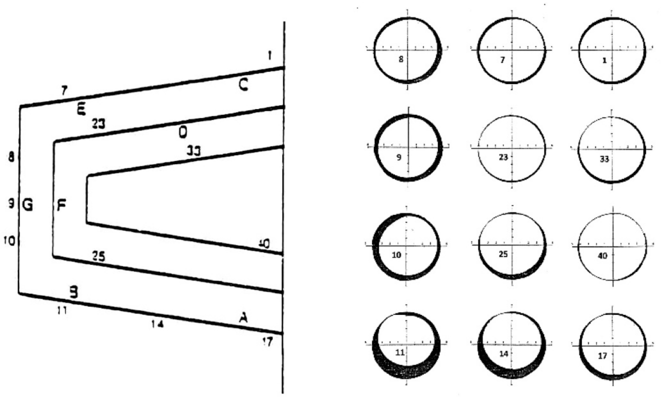


Fig. 4. Material loss in a wing-tube bundle immersed in the bubbling bed of the boiler in Fig. 3d after 1200 h of operation with bituminous coal. The black areas on the tubes represent material loss [32]. The numbers on the tubes refer to the locations on the tube bundle.

movements of the bed. However, a less exposed tube bundle would also be eroded, as it can be seen from the other tubes, which were only slightly affected after 1200 h of operation. Even for more protected tubes, considering that a year has 8760 h and that the tubes should last for many years of operation, the rates or erosion were not acceptable.

On the contrary, no erosion was observed after 3000 h operation in Renfrew [28]. This had also been emphasized by Skinner [33], who based his conclusions on the extensive tests made in British laboratories, claiming that there was no erosion because “the ash formed…is soft and friable compared to the fused ash resulting from higher temperature combustion processes”.

Despite the different results, the conclusion from several boilers was clear: it is not possible to have tubes in bubbling beds because of the relatively high fluidization velocity employed, about 2 m/s (a high velocity is desired to attain a high surface power, Eq. (6)).

There were other experiences as well that made bubbling FBC less favourable than predicted initially:

- low combustion efficiency with coal. Fine char particles are elutriated from the bed and have too short a time to burn while residing in a hot environment. Fly-ash recirculation to a particular part of the bed, called “carbon burn-up cell”, was tried to improve burn up, but in retrospect, it seems more relevant to take the full step to a CFB boiler.
- the sulphur capture was not as good as expected. Limestone particles added to the bed for sulphur capture are also subject to elutriation, but elutriation is not the main cause for inefficient sulphur capture in BFB. The main disadvantage is that the limestone particles are part of the particle phase of the bed, which tends to be under reducing conditions while most oxygen passes the bed through the bubbles. If oxygen is present, SO_2 originating from combustion, is bound by CaO to form $CaSO_4$ but under reducing conditions $CaSO_4$ is not formed. Even if $CaSO_4$ were formed in an oxidizing region (e.g., in the vicinity of bubbles or in the splash zone above the dense bed) it could be reconverted to CaO in a reducing environment, and SO_2 would be released. As a result of insufficient availability of O_2, sulphur capture is far from stoichiometric.
- scale-up to electric utility size led to unfavourable designs with very large bed surfaces because of the low surface power [21]. Moreover, injection of fuel in a dense bubbling bed requited many fuel-feed points. Initially, one feed point per m^2 bed surface area was mentioned. In later designs for biomass, the fuel is injected on the top of the bed, leading to less feed points. Large-scale CFB boilers are designed with in the order of 20 m^2 bed surface area per feed point.
- load control is not without complications. It is easier in CFB where the primary air supply and the ratio of primary to secondary air supply can be adjusted to affect the suspended particle density and thereby heat transfer and load. In BFB two methods have been tried. 1) Bed inactivation, called “bed slumping”, meaning that a part of the bed was defluidized by cutting the corresponding air supply. However, in this case the boundary of the slumped bed tends to sinter because of ongoing reactions without cooling as a consequence of the insufficient transport of heat from the stagnant bed. The buildup of fences to separate parts of the bed results in complications of the normal operation. 2) Change of bed inventory, leaving some cooling tubes outside of the bed to reduce heat extraction from the bed during a reduction of load. This is a more attractive way of load change if erosion on the tubes could be avoided. In the figure showing a pressurized FBC, Fig. 13, such an arrangement is included in the form of a bed material container located in the pressure tank. This method was also applied in atmospheric BFB.

Initially, the interest was on the combustion of coal, but after a few decades, other fuels, such as biomass and wastes were also found to be a useful feedstock for FBC. The experience revealed that coal and high volatile biomass and wastes perform differently: the fuel particles are contained in the particle phase of a bed. There, the fuel devolatilizes, and the char burns if reactants are available. However, the volatiles do not burn in the particle phase of a bubbling bed because the radicals involved in the reactions are quenched on the surfaces of the bed particles [34]. They may burn in bubbles, but they are more likely to burn above the bed. An example of the behaviour of different fuels is given by observations in the Chalmers 16 MW_{th} BFB boiler (Fig. 3d), illustrated in Fig. 5.

Fig. 5 shows the tendency of heat release in the bed and of the cooling of the gases in a freeboard surrounded by boiler tube-walls. The temperature field is rather smooth compared to flame combustion but depends on the size and volatiles content of the fuel. With bituminous coal the profiles are quite even. In the lignite case there is a weak tendency to over-bed combustion, which beomes more pronounced in the biomass case. Particularly the upper curves show that fuel mixing takes place faster than combustion in the coal case, yielding even profiles, whereas in the biomass case fast release and combustion of volatiles occur before the fuel is well mixed. The ratio of the rates of mixing and reaction is fundamental in FBC boiler design and can be analysed in terms of the Damköhler number [36,37].

$$\mathrm{Da} = \text{rate of reaction}/\text{rate of transport}$$

The conclusion made from the experiences gained during the 1970s and early 1980s is that the bubbling bed is not suitable for coal. However, the principle drawbacks of bubbling beds listed above are not valid for biomass if erosion on heat transfer tubes can be avoided (high volatile content and high-reactive char, no sulphur, and limited size of biomass and waste boilers because of cost of fuel-transport).

The approach used for the high-volatile, reactive fuels is to avoid heat-transfer tubes in the bed by starving the combustion in the bubbling bed using staged combustion, namely reducing the air supply to the bottom air-distributor below the stoichiometric amount of air and and adding the remaining air for complete combustion stepwise in the freeboard to avoid locally high temperatures that could easily occur in a burning gas devoid of particles. The char (a minor part of the fuel) burns in the bottom bed and the volatiles in the freeboard.

A generic bubbling FB boiler, shown in Fig. 6, has a refractory-lined bottom part. There are no tubes in the bed, but membrane tube-walls surround the freeboard. Secondary air is added through nozzles in one or several tiers. Superheater tube-bundles may be present in the upper freeboard, as seen in the figure. In addition, there are further superheaters, economizers and air preheaters in the backpass, like in any boiler. Such boilers are quite common in industrial and district-heating applications using biomass and other low heating-value fuels.

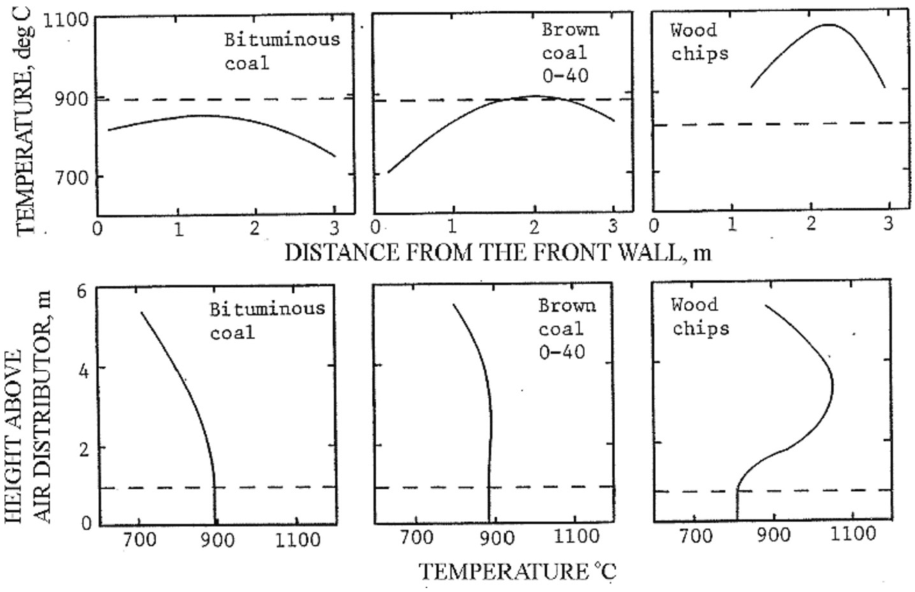


Fig. 5. Upper three diagrams show horizontal temperature profiles for three fuels: coal, lignite, and biomass in the freeboard of a bubbling bed. The lower three diagrams indicate the corresponding vertical profiles. The dashed lines denote bed temperature (upper diagrams) and bed height (lower diagrams). Measurements in the BFB boiler of Fig. 3d [35].

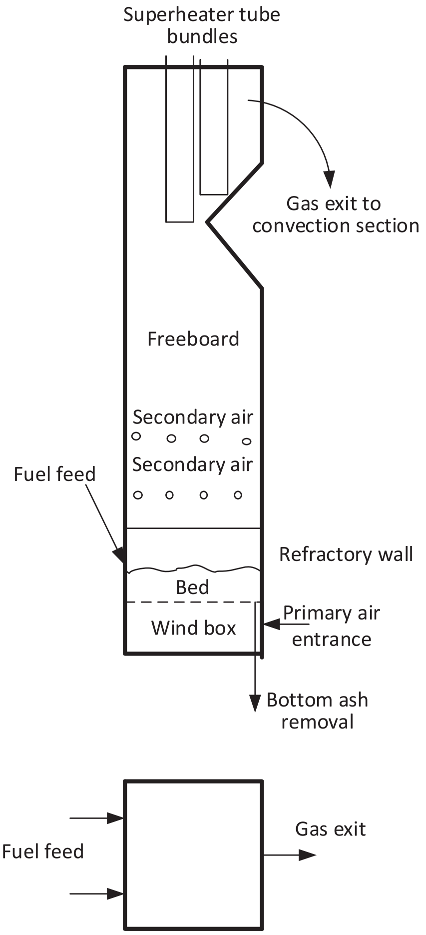


Fig. 6. A generic bubbling bed boiler for biomass and other high-volatile fuels. The sizes of such boilers are in the order of 10 to 100 MW_{th}.

3.3. *Circulating FB combustors (CFB)*

3.3.1. *General principle of CFB*

Catalytic reactors are operated with catalyst particles of narrow size distributions. Boilers, on the other hand, are charged with a fuel that contains ash, and the resulting bed will have a wide size distribution. The larger particles tend to remain in the bubbling bottom bed, while the finer ones can be carried away by the gas. This fine fraction is a loss unless it is captured by a particle separator (usually a cyclone) and returned to the bed. This was realized already by Winkler, Fig. 1b.

The inconveniences experienced while operating the BFB boilers are reduced by increasing the fluidization velocity and recirculating the elutriated bed material through a particle separator and so increasing their residence time and the efficiency of conversion of char and limestone for sulphur capture. Moreover, the increased specific surface-power (Eq. (6)) facilitates scale-up, and the entrainment of bed particles carries heat to more protected heat-transfer surfaces. Consequently, scale-up to reasonable sizes for electric utility operation is more convenient at a high velocity than with low-velocity beds.

The high velocity gas entrains particles to the upper part of the furnace where additional heat transfer surfaces receive the heat produced by combustion in the lower bed. Heat transfer is mostly radiative and the contact between particles and surface is moderate, hence, erosion is low at velocities below 6 m/s. Circulation as such is usually not a goal but rather a consequence of the higher velocity. Therefore, the rate of circulation is not important. However, there are exceptions to this statement:

- larger boilers may have external heat exchangers. Then, of course, the rate of particle recirculation to these heat exchangers determines their capacity.
- combined reactors (see dual FB, below). In such devices, the transport of particles from one reactor to another controls the operation.
- there are fuel converters where the interaction between bed particles and fuel is important (for instance, chemical looping combustion). A plug-flow-like situation is desired. Then the gas-particle interaction controls the conversion, and the concentration of particles along the height of the reactor has a significance like that in a catalytic reactor.

With a circulation flux of particles of the i-th size fraction, G_i (kg/m^2, s), in each circulation turn the amount $G_i(1-\eta_i)$ is lost (Fig. 7), controlled by the fractional efficiency of the separator η_i. Together, the input flux will get lost after n_i turns when the flux is $G_i = n_iG_i(1-\eta_i)$ and from there the number of turns, n, through the circulation loop is given, as shown in Eq. (7).

$$n_i = 1/(1-\eta_i) \tag{7}$$

Further modelling is required to evaluate the residence time of reacting particles.

Clearly, the separator efficiency η_i is another important factor for the performance of a circulating bed. This efficiency determines the quantity of particles circulating under the condition that they can be carried by the gas.

The lower curves in Fig. 8 illustrate the sizes of the circulating particles in a CFB boiler. Both the circulation rate G_i and the size $d_{p,exit}$ of the particles carried away by the gas to the furnace exit increase with fluidization velocity, while the bulk of the bed retains more or less its size; only attrited particles, ash, and char are produced as small circulated particles. It is noted that the bed circulates even at low velocities when the circulated particle size is comparatively small as well as the circulation rate. So, when should a bed be called "circulating bed"? When the transport velocity [39] is attained? It seems, if the transport velocity, based on the size of the bed material, is taken as a criterion, this circulating fluidized bed is never circulating because the transport velocity is found above the fluidization velocity of the bed, somewhere between 7 and 9 m/s (depending on correlation used). Even if the concept "CFB" is clearly defined, the application of the concept is not straightforward in all cases. Here it is proposed, simply, that if particles are captured from the gas and recirculated to the bed the device is circulating, irrespective of gas velocity and rate of circulation, even though the bottom bed is still bubbling. In fact, it has been shown that a normal CFB boiler is operating with a bubbling bottom bed (although the bubble shape in this case is irregular because of the high velocity) [40]. This deviates from the generally accepted concept (inherited from chemical reactors) that CFB requires a high gas velocity and a high circulation rate, and that the bed is "fast fluidized".

Researchers from the Tsinghua University in China have developed a similar idea, subdividing the bed content into one "ineffective" part, mostly forming the bottom bed, and one "effective" part forming the circulating particles. By careful fuel preparation and improving the efficiency of the cyclone, the ineffective part can be reduced while the effective part remains. This leads to several advantages for the performance of a CFB boiler, summarized in [41]. Measurements presented [41] show circulating particle sizes in the range of 0.1 to 0.2 mm at full load (4 to 6 m/s), in contrast to the particle sizes of Fig. 8 that reach 0.3 mm at 6 m/s. Further measurements are needed to explain the different particle sizes in the two cases, but some differences between the operation situations in the two cases can be pointed out: The tests in Fig. 8 were carried out with all air from the bottom of the furnace, while the Tsinghua data most likely are taken during normal operation of CFB boilers, namely with a certain separation of the air supply between primary and secondary air. This leads to different entrainment rates and presumably also to different size distributions. Unknown differences in the cyclone efficiencies may also have contributed.

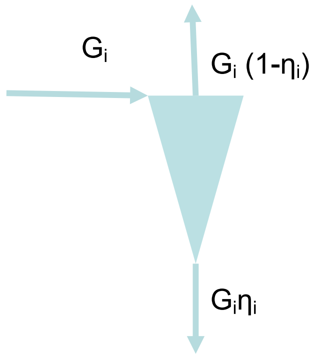


Fig. 7. Particle separator (here represented by a cyclone) with particle fluxes G_i for a separator efficiency η_i of the size fraction i.

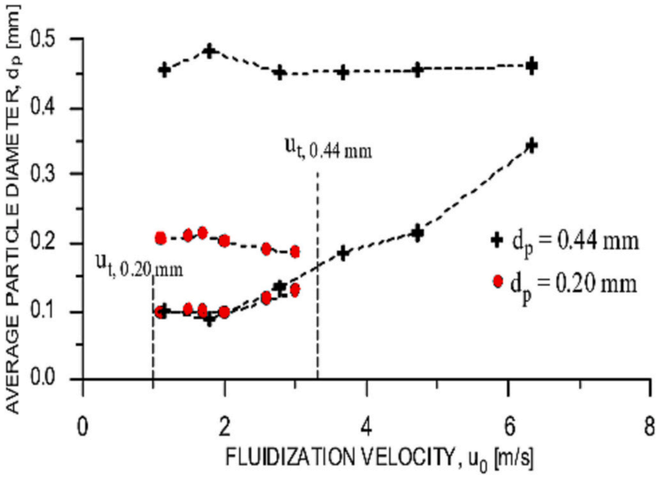


Fig. 8. The particle size distribution in a CFB bottom bed (upper curves, $d_{p,bed} = 0.44$ mm and in another case 0.20 mm). The lower curves show the size of the circulating particles $d_{p,exit}$ measured in the return leg of the CFB boiler at different fluidization velocities during the combustion of coal (only primary air was used in this experiment) [38].

The generally accepted concept of a circulating fluidized bed was not introduced early, but to discuss the development a definition is needed. Goral et al. [42] forwarded the assumption that this concept was introduced by the applicants of the first Lurgi patent on a CFB boiler [43,44] Reh, Hirsch, Collin, and Flink, during a discussion regarding their patent in 1975. However, this cannot be true, because Reh had already used the denomination (in German) in an article 1970 [45]. Alternatively, it was often called "fast FB", but this is a regime of fluidization introduced by Yerushalmi et al. [46], borrowed by the vendors of CFB when introducing the new design to distinguish it from "bubbling" FB.

Looking outside of the field of solid-fuel conversion, circulating beds were an obvious solution in the petroleum industry where circulation of bed particles took place between a catalytic reactor and a regenerator and back again. It was "invented" because of the desire to increase the fluidization velocities to enhance conversion in a catalytic bed. The bed particles tended to be blown away at increased velocities, and it was a reasonable action to try to recirculate them using a cyclone, Lewis [47]. However, a circulation system was proposed already by Winkler (Fig. 1b), and it was introduced "spontaneously" to improve the combustion efficiency in Fig. 3b, showing a stationary (bubbling) FB with recirculation of particles to the bed. With an efficient cyclone, a considerable circulation can be achieved, such as was the case in the improved HTW gasifier.

The closest predecessor to CFB boilers is found in the calciners developed by Lurgi. They were put into operation for calcination very early. Examples are seen in Fig. 9, presented by Reh in a lecture the year 1969 and then published [45], and in patent applications, Schytil [48].

The reactor of this calciner had a diameter of 1 m and a height of 8 m. The walls were insulated, and it was fired with oil to supply the heat for the calcination reaction. Otherwise, it has the features of subsequent CFB boilers—the riser, the cyclone, the loop seal, and an external heat exchanger, serving to cool the calcined material.

The step from calciners to boilers was taken, inspired by Swedish inventors, who first wanted to develop an iron-reduction process (Flink, working in a mining company and Collin from the company ASEA, later ABB) and Lurgi was contacted to support the design of a gasifier [42].

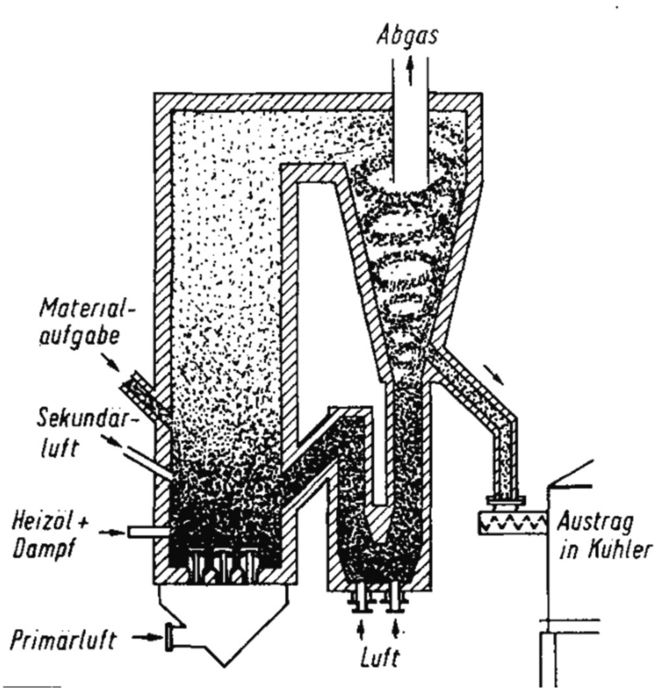


Fig. 9. Calciner operating from 1970 in Germany. Particles are extracted from the cyclone to an external particle cooler [45].

This process (Elred) was not commercially successful, and the same inventors (Flink and Collin) together with representatives from Lurgi (Reh and Hirsch) went on to develop a CFB boiler, in the first place to burn Swedish oil-shale to extract uranium, but they formulated their patent in a general way to be valid for carbonaceous fuels [43,44], and so they covered all important applications within the field of combustion of solid fuels. Lurgi's version of the development is presented in some detail by Goral et al. [42] where they also explain that their patent became dominant and other manufacturers, developing CFB boilers, had to make patent agreements with Lurgi. This is surprising because, as shown by Leckner [21], at the same time there were several other patents regarding CFB boilers, for instance [49,50]. The Lurgi patents included water walls in the furnace as is typical for a boiler, but nevertheless, the first boilers produced by Lurgi, Lünen [25] and Duisburg [51], looked more like calciners with a mostly refractory-lined combustor connected to an external heat exchanger. Steven [52] mentions that 30% of the useful heat was transferred in the furnace and the back pass, the rest went to external heat-exchangers and losses. A similar approach was shown in several of the early patent applications [49,50]. Initially, many designs were proposed, but after some decades of development, with the early patents expired, the form of a CFB boiler converged into a rather common shape, just like the automobiles: the general aspect is similar for various brands, the differences between manufacturers are in the details.

3.3.2. Description of CFB combustors

Many manufacturers took up the design of CFB boilers in the beginning of the 1980s (some years after the first patents). The further development consisted in the refinement of the technology and a gradual increase in the maximum size from a few tenths of MW_{th} to almost 2000 MW_{th} (somewhat above 600 MW_e) in the 2020s.

Most CFB boilers converged into the generalized form shown in Fig. 10 with only minor individual differences. Typical features are:

- Tapered bottom walls;
- Furnace walls covered by membrane, evaporator tubes;
- Additional internal heat-exchanger surfaces, including superheaters;

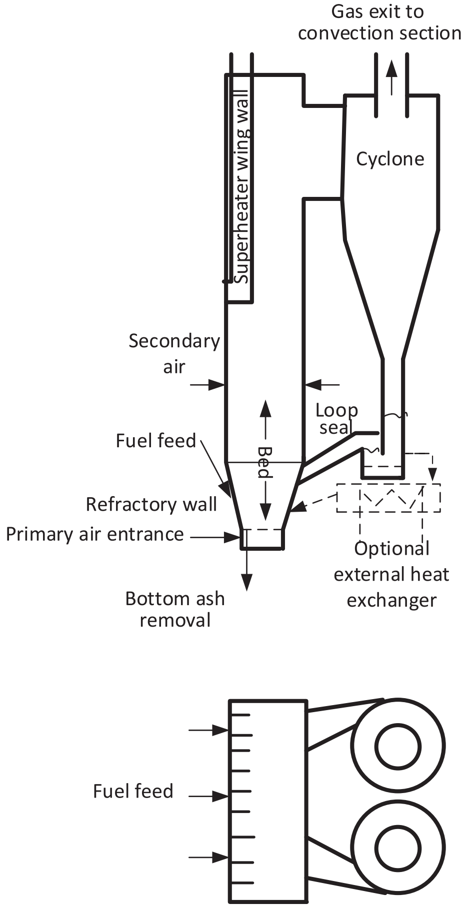


Fig. 10. A general shape of a CFB boiler.

- Cyclone (both cooled and uncooled);
- Loop seal;
- Primary/secondary air;
- Rectangular cross section.

Typical furnace dimensions including cyclones are presented on Fig. 11.

The furnace is tall but smaller than that of a pulverized fuel boiler, Fig. 11. The cross section grows rectangular as the size increases because

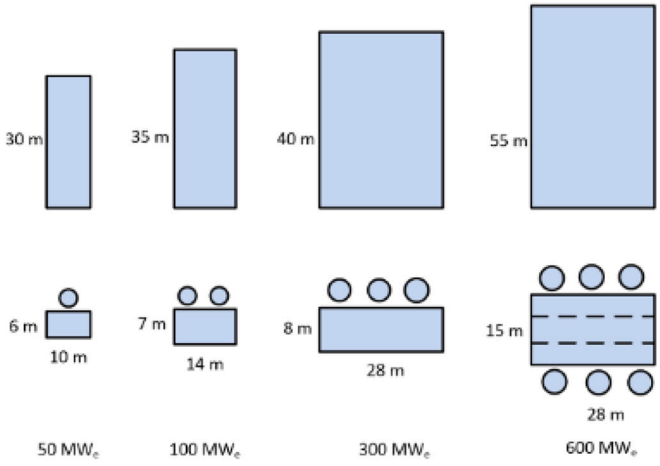


Fig. 11. Dimensions of CFB furnaces of different sizes equipped with several cyclones.

air and fuel are introduced from the side and mixing over the entire cross section is important. Primary and secondary (sometimes also tertiary) air is used. The bottom walls of the furnace are usually tapered and refractory lined. The primary air is introduced through a water-cooled bottom distributor plate consisting of many nozzles and including some arrangement to extract bottom ash. There are several cyclones, because of space limitations and because the efficiency of a cyclone falls with increasing size. The maximum size is usually below 10 m in barrel diameter. To avoid very long rectangular furnace sections, the bottom part of the largest boilers is often divided into two legs, such as indicated for the 600 MW_e design in Fig. 11. The very large boilers must have cyclones on both side walls, something that reduces the space for heat extracting wing walls. Then, internal walls extending from bottom to top and external heat exchangers are required to provide sufficient cooling surface.

The allocation of the heat transfer surfaces is critical. They should be inserted to avoid erosion. First, there are furnace-internal heat transfer surfaces, usually membrane-tube walls and wing walls in the upper part of the furnace, aligned in parallel to the main direction of the particle flow. Second, like in all boilers, there is a back-pass with superheaters, economizers, and air-preheater surfaces. Finally, in some designs (usually larger than 300 MW_e) with six or more cyclones external heat exchangers extract heat from the recirculating particles. Fig. 12 gives a survey of different locations of internal heat-transfer surfaces in the loop of a CFB (a small heat exchanger placed in connection with the loop seal is also internal, in contact with the circulating loop. An external heat exchanger is seen in Fig. 10.).

3.4. *Efficiency enhancement*

It is a general trend in the production of electric power to search for means to reach high (optimized) efficiencies. The efficiency is related to fuel, size of plant, and type of energy system (electric utility, industrial, waste treatment, district heating). The electric power plants concerned are almost always based on a Rankine cycle with steam as a working medium. In rare occasions combined cycles Rankine-Brayton have been applied (IGCC, steam-gas-turbine cycles, combined cycles). Starting hundred years ago, pulverized coal (PC) combustion has grown to be a principal means of solid fuel conversion. Table 3 gives an overview on efficiencies with coal or waste as a fuel.

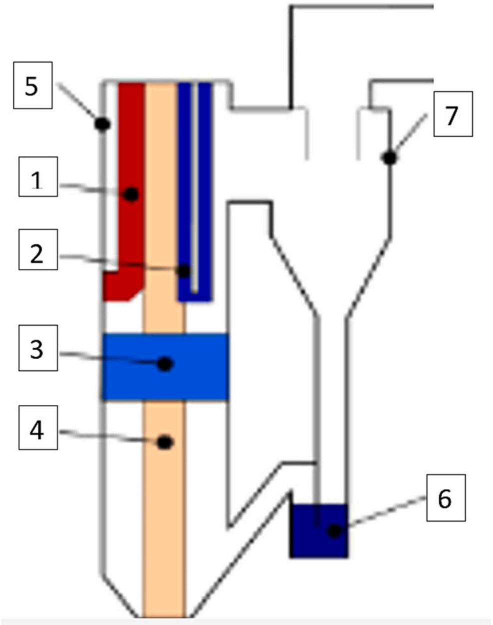


Fig. 12. Heat transfer surfaces in a CFB furnace, 1) Wing walls; 2) Hanging superheater;3) "Omega" tubes (not common due to cost); 4) Inner panels extending from bottom to top; 5) Membrane tubes on the furnace walls 6) Furnace external heat-exchangers, in this figure the heat exchanger is integrated in the loop seal. 7) Steam or water-cooled cyclone. All surfaces are protected against erosion in critical locations. (After Goidich, Foster Wheeler [54]).

Combined cycles can be found in the upper ranges of efficiency or even higher than those mentioned.

In the waste (MSW) conversion area, grate firing is so far the most common combustion method used in practice, but FBC has advantages in a better controlled combustion, permitting less excess air and higher steam data. Moreover, FB combustion allows easier change of fuel from one waste to another or to combinations of fuels, co-combustion. The drawback of FB compared to grate-firing is in the need for fuel preparation, sorting out metals, glass, and other large objects [53].

Among sub-critical boilers the difference between FBC and its main competitor PC is in the fuel: a PC boiler uses ground fuel and then certain restrictions on the quality are imposed, whereas FBC can handle more irregular fuels with less requirements on quality. In the supercritical ranges of steam data, FBC has advantages in front of PC: The heat-load profile on the furnace walls is more even in a CFB boiler than in PC that has a distinct impact on heat flow from the hot flames in certain regions of a furnace. This means that less expensive heat-transfer tubes (without heat-transfer enhancement on the steam-water side) and smaller water flow in the tubes can be used in supercritical designs of CFB boilers [54]. There is a limitation to extreme steam temperatures, though, because of the usual maximum temperature on the furnace side in a CFB boiler of 900 °C, while a much higher temperature is available in a PC boiler.

Another option to reach high efficiencies is the combined cycle. As mentioned above, the HTW gasifier was developed for high pressures to deliver gas to the combustion chamber of a gas turbine whose exhaust gas would be utilized for a steam cycle. This proposal to apply HTW was not successful because the German power companies preferred a power station based on a single Rankine cycle with high steam data burning natural gas, capable of reaching even higher efficiencies than those of Table 3, which are for coal. Besides, in the intended large-scale application, the FB gasifier has a serious competitor in the entrained-flow gasifiers.

With this background, the idea forwarded in the UK already in the 1960s [55,56], and subsequently investigated in several research laboratories, was genial, namely, to combine the gas turbine combustion chamber with the steam boiler of the Rankine cycle in one pressurized unit, Fig. 13.

The compressor of the gas turbine supplies the air for fluidization and combustion in the fluidized bed. According to Eq. (6) the high pressure yields a high specific heat release, and the fluidization velocity can be chosen low in the bubbling FB to mitigate erosion on the densely spaced cooling tubes for evaporation and superheating of steam in the bed, while still maintaining an acceptable surface power. Limestone could be supplied to the bed for sulphur capture.

The right-hand figure shows the bed inside its pressure vessel with cyclones for coarse cleaning of the exit gas. The bed height that could reach four metres at full load is reduced to allow smaller heat extraction at lower loads, keeping the bed temperature constant. The storage container for bed material, used during load changes, is shown inside the pressure vessel. The flue gas passes a filter before it enters the gas

Table 3
Order of magnitude steam data and efficiencies for various electric power plants operating with solid fuels.

Type	Steam data: pressure, MPa, temperature/ reheat, °C.	Typical net efficiency, %
Waste combustion (MSW)	Subcritical, 8, 400	25
Utility	Subcritical, 17, 540/540	39
Utility	Supercritical. 25, 570/570	43
Utility	Ultra-supercritical, 28, 600/600	45

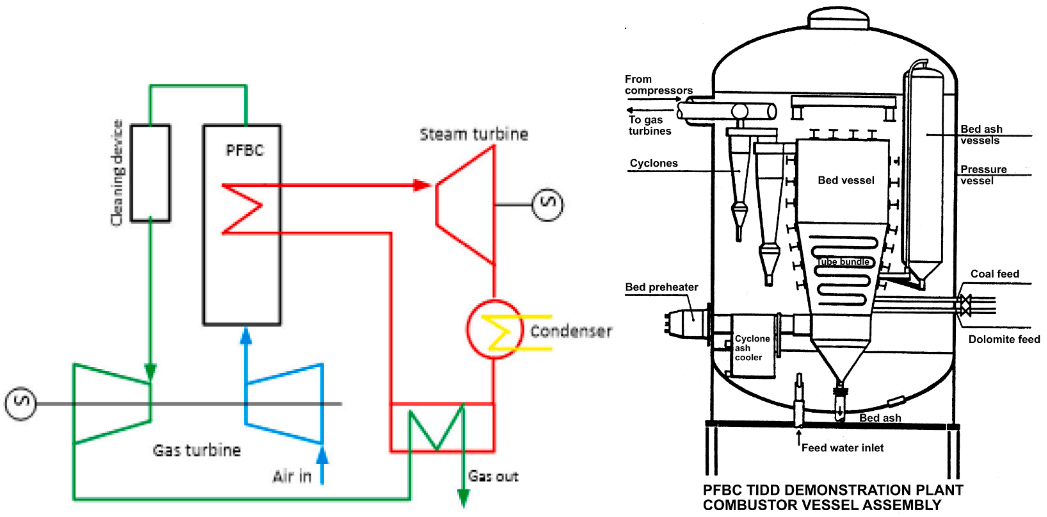


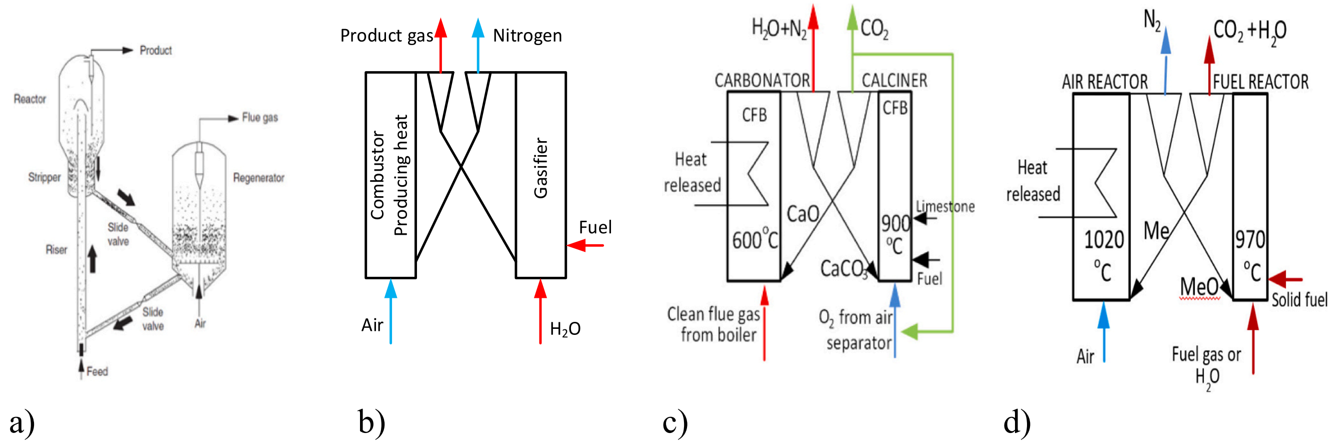
Fig. 13. Scheme of a PFBC process with a pressurized FB combustor (PFBC) presented in the right-hand figure.

turbine. This filter is a weak point in the system, and much work was spent on the development of ceramic candle filters, granular-bed filters, and others, as they should operate at bed temperatures of 800 to 900 °C, a very high temperature from the point of view of filter materials. At the same time, this gas temperature was a serious limitation of the process since modern gas turbines can handle much higher temperatures of about 1500 °C. Therefore, efforts were made to solve this problem, and a "second generation" process was proposed by Foster Wheeler [57]. This pressurized process first introduced the fuel in an FB "carbonizer" where it was devolatilized and possibly mildly gasified. After cleaning, the gases went to the gas turbine's combustor to boost the temperature of the gas from the main combustor, where the char was burned at high excess air. The bed material of the CFB was circulated to an external FB heat exchanger (a "dual-reactor system", see below) where the steam for the steam cycle was produced, added to the steam from a boiler, heated by the exhaust gas of the gas turbine. It was estimated that the plant efficiency would be about 45% (based on the higher heating value of the fuel). Around the change of the millennium, a plant was to be erected at Lakeland, Florida USA, but afterwards nothing was heard of the fate of the project nor about the process as such. The same destiny seems to have resulted in the disappearance of other pressurized FB plants. No activity in the area has been recorded recently. Further information on PFBC and particularly about the development in Japan has been published by Shimizu [58].

4. Dual and multi-reactor systems

Catalytic reactors were employed for cracking in the oil industry, but the catalysts required regeneration to remove deposits. It was natural to establish a dual-reactor system, consisting of a catalytic reactor and a regenerator coupled to each other, Fig. 14a).

Here, fluidized beds replaced fixed beds, and to enhance the rates of reaction it was reasonable to increase the fluidization velocity to connect the catalytic reactor with the regenerator through cyclones. It was a great achievement when the first Fluid-bed Catalytic Cracker (FCC) was put into operation in the Baton Rouge refinery (USA) in 1942 [59]. It took some years before other areas than the oil industry took advantage of the new concept. In Japan, Kunii and coworkers [60] had developed a two-reactor process, the KK reactor for the petroleum industry [2]. This device later inspired the design of a waste gasifier for 40 t/h of shredded MSW, based on fluidization with two coupled reactors, put into operation outside Tokyo [2]. The operation is reported to have been successful [61]. Despite that, later, Kunii and coworkers [62] expressed the opinion that twin-reactor systems were complicated and that it would be better



a) b) c) d)

Fig. 14. Examples of coupled FB reactors. a) Fluid-bed catalytic cracker for petroleum treatment b) Allothermal gasifier c) Calcium looping system d) Chemical looping system.

to apply the process as an internally circulating device. In contrast to Kunii, Hofbauer at the TU Vienna started with an internally circulated bed, described in his Dr-Thesis 1982 and later in the SCEJ Symposium [63], but he ended up with a twin-FB reactor, a first version of which is described by Hofbauer et al. [64]. A slightly more developed version of this concept, consisting of a coupled gasifier-combustor to produce heat for a biomass gasifier, was successfully operated in Güssing, Austria, and then introduced in several Austrian and German district-heating systems for production of electric power by gas engines, while the exhaust heat from the engines was utilized for heating of buildings. A late version, Fig. 15, was delivered to the district heating system of Göteborg, Sweden, to produce substitute natural gas and heat from biomass. The project was called Gobigas.

Fig. 15 shows the bubbling bed gasifier, which is actually a devolatilizer, connected to a CFB riser for combustion of the char that results from the gasifier, and a connected cyclone to recirculate the hot bed material to the gasifier.

The gasifier system is simple, but tar is always produced in biomass conversion at low temperature (800–900 °C) and must be removed from the product gas. The remaining char should be sufficient for the heating of the gasifier bed and the fuel fed. Otherwise complicating measures must be taken, such as to feed additional fuel to the combustor. If the gas is to be used to produce methane like in Göteborg, several further process steps are required: a shift reactor, a reformer, CO_2 removal from the gas, guard reactors to avoid contamination of the catalysts employed, in all, something between 15 and 20 reactors [65].

The purpose of the Gobigas plant was to produce 20 MW_{fuel} of biomethane, to determine the performance of a plant, and to acquire experience for the design of a larger unit of >100 MW_{fuel}, deemed to be profitable. The 20 MW_{fuel} plant was put into operation, worked well, and gave the intended valuable information for scale-up [66]. However, in the meantime there was a change in the political configuration in the commune of Gothenburg and the new leadership decided to close the plant with the motivation that it is not the task of the taxpayers of Gothenburg to solve the greenhouse gas problem. This is the destiny of many biomass FB gasification projects. Still, the fossil fuels are too cheap.

The devices in Fig. 14 are drawn as simplified CFB but they could be BFB also, Fig. 14a) and b) have been commented above. The two remaining examples 14c) and d) are removing CO_2 from nitrogen-containing flue-gases for compression and sequestration. Fig. 14c) separates CO_2 from the flue gas of an air-fired boiler by addition of lime, and d) uses a metal powder for oxygen transport by oxidizing it in an air reactor and reducing it in a fuel reactor. a) is widely applied in refineries, b) exists in a few cases (Fig. 15), whereas c) and d) are only proposals being investigated up to pilot scale.

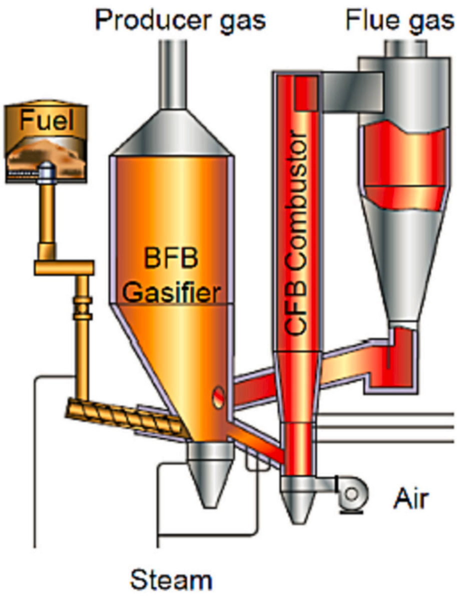


Fig. 15. A dual FB reactor of Güssing type delivered to Göteborg.

The purpose of the coupled FB reactors shown in Fig. 14 is to separate the tasks performed in each reactor, using the same particulate material circulating between them, and also to separate the effluent gases as summarized in Table 4.

For instance, in the gasification unit described in Figs. 14b and 15, the heat needed for the process is provided by combustion of a part of the fuel in one of the reactors. Then the hot bed particles are transported to the other reactor to heat the fuel there, where devolatilization and gasification take place. In this way the product gas leaves the gasifier without being diluted by the air nitrogen from combustion that leaves from the combustor. In a single-reactor gasifier, combustion and gasification take place in the same reactor and, unless oxygen is used for combustion, the nitrogen from the air will dilute the product gas, reducing its heating value.

5. Discussion on the influence of environmental conditions on the technical development

During the first 80 years, coal was the target fuel for FBC. After that, gradually, the awareness increased of the global warming caused by the conversion of fossil fuels, and other solutions are looked for. The environmental issues have always been in the foreground for the development of FBC after the Winkler period. The reason why FBC became of interest in the UK and USA was the increasing oil prices, giving coal an advantage, but also the finding that high NO emissions could be avoided and that sulphur capture by limestone addition to the fluidized bed was possible, thus avoiding expensive flue-gas cleaning. The knowledge on the emissions of harmful substances increased further since then and is quite mature at this moment. It also became clear that the FB is capable of handling various fuels like residual coals, wastes, and biomass in single mode conversion or in co-conversion, that is, using mixtures of fuel. Lately, the expectations on biomass have increased excessively. It is likely that this limited resource will not be sufficient to satisfy the demand.

The alternative route to the reduction of greenhouse-gas emission would be to continue to use coal, by capturing and disposing of the CO_2, CCS (Carbon Capture and Storage). FBC is very well adapted to several of the proposed routes for CO_2 capture: preconversion, postconversion, and oxyfuel conversion. *Preconversion* implies to first gasify the coal, transform it into H_2 to be burnt in the combustion chamber of a gas

Table 4
Examples of dual FB reactor systems and their products.

Application	Reactions	Gases	Solids
Catalytic reaction such as heavy oil cracking	Cracking with deposit formation / Catalyst reactivation by deposit burning	Light oil and fuel gas / Flue gas (CO_2)	Catalyst
Air-blown autothermal biomass gasification for medium heating-value gas	Biomass gasification (endothermic)/ Char combustion (exothermic)	Syngas (CO, H_2) / Flue gas (CO_2)	Inert material (heat carrier)
Calcium-looping CO_2 separation	CO_2 capture by CaO from flue gas/ CO_2 release from $CaCO_3$	Remaining flue gas / Concentrated CO_2	CaO/$CaCO_3$
Chemical-looping combustion	Reduction of metal oxide by fuel/ Oxidation of a reduced metal oxide by air	Concentrated CO_2 / Oxygen-depleted air	Metal oxides (e.g. ilmenite, or mining wastes)

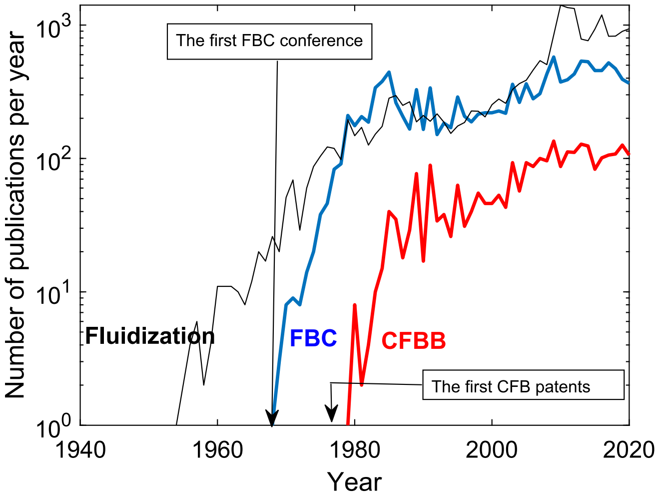


Fig. 16. Number of publications per year dealing with Fluidization, FBC or CFBB (from Scopus).

turbine or converted to electricity using fuel cells after the removal of the CO_2 from the gas. This is suitable in large-scale units where probably FB gasification is less competitive compared to entrained-flow gasifiers, except for high-ash, reactive fuels. *Postcombustion* can be carried out in a calcium-looping system where the CO_2 from the flue gas of a conventional boiler enters an FB reactor, the carbonator, fed with CaO to absorb the CO_2. The $CaCO_3$ formed is then recycled to an FB regenerator, a calciner, to be reconverted to CaO and recycled to the carbonator. The arrangement is a twin-bed system like those shown in Fig. 14c. There are two kinds of *oxyfuel* combustion depending on how the oxygen is produced. In one option the oxygen is added from an outside source, for instance by cryogenic air-separation, and the combustion takes place in a conventional FB (or even a pulverized fuel) combustor. In the other option it is produced inherently in the process, in a CFB air reactor, by oxidizing an oxygen carrier (a metal powder), which is then recirculated to an FB fuel reactor where it is reduced to form CO_2 and H_2O, thus

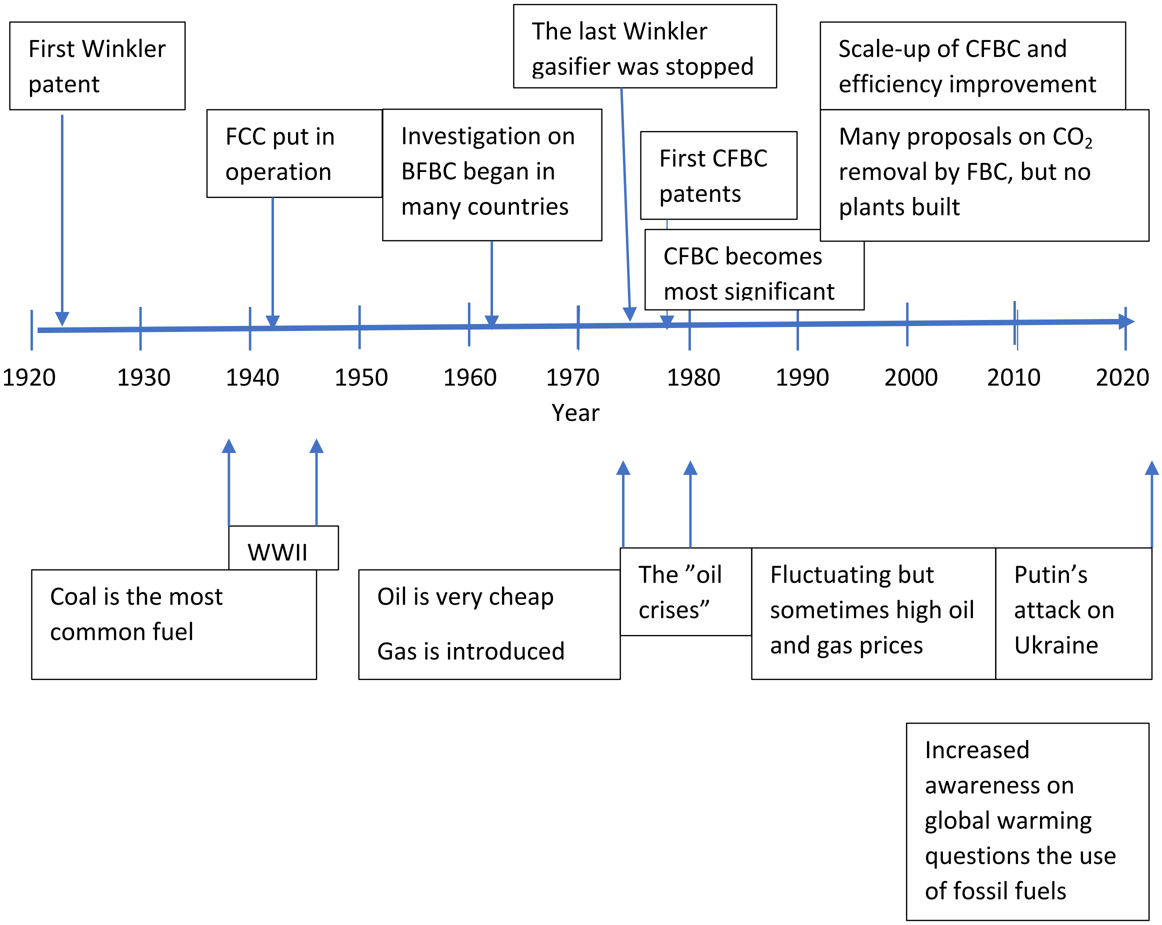


Fig. 17. The 100 years of development of FBC (upper part) contrasted by some aspects of the influencing background situation (lower part).

consuming the fuel. This dual reactor system is called Chemical Looping Combustion. So far, none of these methods to capture CO_2 has been applied commercially.

The introduction of natural energy in the forms of wind and solar power with their time variations are imposing a new challenge on FB combustors when these contributions grow important in an energy system if adequate storage methods are not available. Load following may be required. This can be done with available knowledge but improvements and refinements to adapt operation to new situations of load change should be investigated.

6. Trends in publications on FB fuel conversion technology

The number of scientific publications on a particular topic can be taken as an indication of the interest in this topic. Fig. 16 shows the number of publications per year listed in the Scopus database.

It is not clear how efficient the database Scopus is to cover publication before 1950, but it is for sure that the number of publications from that period of time is small and that there is a remarkable growth following the events noted on the diagram: FBC arouse a considerable interest at a time coinciding with the first FBC conference (or rather, the first conference was held because of the considerable interest emerging at the time). The subsequent interest in CFB is quite logical. The newly issued patents and the rising awareness of the advantages of CFB compared to the previous BFB, as has been pointed out above, explain the exploding interest. Fluidization is a more general topic, and it is understandable that the corresponding curve starts earlier than the publications related to solid fuel three curves; just after the year 2000 there is a marked increase in the number of publications. A closer look at the statistics reveals that this increase is caused by an expansion in publications from China, where interest in fluidized bed combustion has increased notably, although, as mentioned above, China started quite early. Besides, the statistics are based on the classification of the authors, and that explains why the number of publications related to FBC some years exceeds those having the keyword Fluidization.

7. Summary and conclusions

A final overview of the most important development steps accompanied by some influencing factors from the surrounding global environment, notably the energy prices, are presented in the diagram of Fig. 17.

The Winkler gasifier, introduced in a patent application 100 years ago, was successfully used in Germany till the 1970s and was later improved and further developed in the form of the High-Temperature Winkler gasifier, first to be used in chemical industry and then proposed for application in a combined-cycle power plant. Subsequently, it was proposed for conversion of biomass and waste. None of these intents of commercialization was successful. Most likely, the reason is the bavailability of cheaper natural gas. Many other FB gasifier designs suffered the same fate.

The concern for the environment was increasing in the 1960 and 70s and arouse a significant interest in FBC, as such devices were expected to yield low emissions of nitrogen oxides, and through addition of lime to the fluidized bed, sulphur could be captured. First the interest was in bubbling FB. Later it was found that circulating FB had several advantages and that type of boiler started to dominate. The initial success of FBC in Europe and USA was followed by a worldwide expansion, particularly in China, and during the first decade of the new millennium commercial plants in sizes above 600 MW_e and with supercritical steam data were introduced. Likewise, there was an expansion of smaller industrial and district heating plants burning biomass and waste.

Recently, the situation changes. The increased awareness of the global warming by conversion of fossil fuels has made many countries hesitate in expanding their utilization of coal and consequently of FBC.

However, during the last two decades many proposals of using FBC for CO_2 capture have been made: precombusteion (gasification), post-combustion (calcium looping etc.) and oxy-fuel (with oxygen from air separation or from oxygen carriers etc.). Those proposals have been studied frequently and a considerable knowledge is available. Now, large-scale plants involving CO_2 capture are required to further forward the technology of FB conversion.

Nomenclature and acronyms

A	reactor cross-section area, m^2.
c_{pm}	mean specific heat, J/kg,K
d	reactor diameter, m.
F	fuel flow, kg/s.
f	factor in Reaction R1.
G	gas flow, m_N^3/h.
H	heating value, J/kg fuel.
g	specific gas quantity, kg/kg fuel.
m	moisture in gas, % of gas flow.
n	number of turns through the circulating loop, −
P	pressure, Pa.
q	heat power removed from a furnace, W/kg fuel.
Q	specific heat load, W/m^2 bed surface.
T	temperature, ^{o}C or K.
u	fluidization velocity, m/s
ρ	density, kg/m^3.
η	efficiency, −.
ψ	factor related to the degree of fuel combustion, −.

Indices

bed	bed
i	size fraction
LHV	lower heating value
o	standard condition
sec	secondary gas

Acronyms

FB, BFB, CFB,	fluidized bed, bubbling FB, circulating FB
CFBB	CFB boiler
CCS	carbon capture and storage
EU	European Union
FBC, PFBC	fluidized bed conversion, pressurized FBC
FCC	fluid-bed catalytic cracker
PC	pulverized coal boiler, combustion
HTW	high-temperature Winkler gasifier
IGCC	integrated gasification combined cycle
MIT	Massachusetts Institute of Technology
SCEJ	Society of Chemical Engineers Japan
TU	Technische Universität
UK	United Kingdom
USA	United States of America
WWII	World War II

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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