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DEVELOPMENTS IN FLUIDIZED BED CONVERSION OF SOLID FUELS

by

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A summary is given on the development of fluidized bed conversion (combustion and gasification) of solid fuels. First, gasification is mentioned, following the line of development from the Winkler gasifier to recent designs. The combustors were initially bubbling beds, which were found unsuitable for combustion of coal because of various drawbacks, but they proved more useful for biomass where these drawbacks were absent. Instead, circulating fluidized bed boilers became the most important coal converters, whose design now is quite mature, and presently the increments in size and efficiency are the most important development tasks. The new modifications of these conversion devices are related to CO_2 capture. Proposed methods with this purpose, involving fluidized bed, are single-reactor systems like oxy-fuel combustion, and dual-reactor systems, including also indirect biomass gasifiers.

Key words: *fluidized bed, combustion, gasification, chemical looping, oxy-fuel, calcium looping*

Introduction

The development of fluidized bed conversion (FBC) during the last hundred years is illustrated in fig. 1, where various lines of evolution are seen.

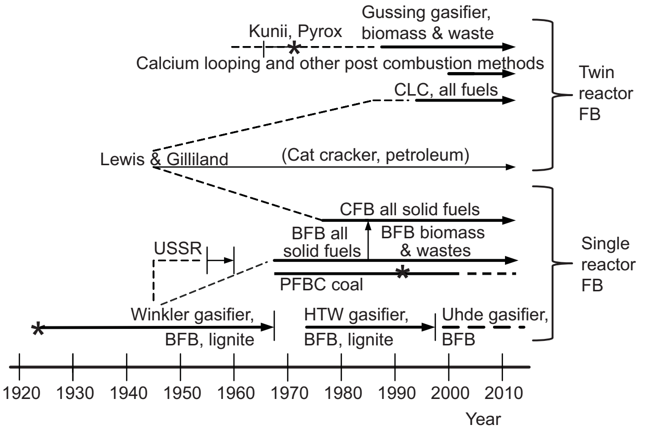

Figure 1. Fluidized bed conversion history. The lines show the main paths of development

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It is the purpose of this article to make a concise survey over the historic period covered by fig. 1, commenting on the various lines of development and their connections.

The Winkler line

The technical development of FBC was first manifested in 1922 in a patent by Winkler [1] on a bubbling fluidized bed gasifier. Almost 40 units of this type were built for production of syngas for the chemical industry, starting 1926 in Leuna, Germany, and ending in 1975. The Winkler gasifier is a "direct gasifier" where some combustion by air or oxygen is performed in the gasifier bed to produce heat for the endothermic gasification reactions and for heating of the fuel and the bed.

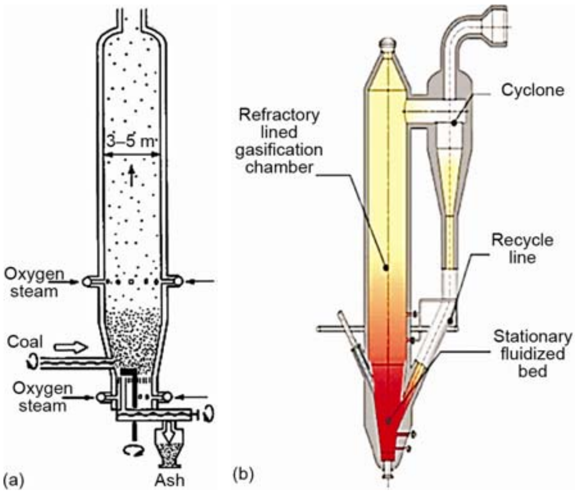

Figure 2. (a) Early Winkler gasifier, (b) modern high-temperature Winkler gasifier (both are "bubbling" beds despite the cyclone)

A drawing of an early version of the gasifier in fig. 2(a) shows a rather large refractory-lined reactor whose cross-section is up to 5 m in diameter, with staged steam/oxygen supply, and an arrangement for extracting the ashes from the bottom. From the drawing it is evident that particles were also carried away with the gas. Well-known drawbacks during operation with coal (lignite) were clinkering, limiting the operation temperature to less than 1040 °C, tar formation at this "low" temperature, and loss of char in the removed bottom and fly ashes.

This type of gasifier was improved in a design called high-temperature Winkler (HTW) gasifier in Germany, shown in fig. 2(b), introduced in 1974 and successfully developed till the year 1997 for applications in power generation and production of methanol [2]. At that year the work was stopped: it was not economical to produce syngas from lignite. A later effort in commercialization aimed at power production in combined cycle, but high-efficiency Rankine cycle with pulverized coal was finally preferred. Recently (2011), the manufacturing rights of the HTW gasifier were taken over by the German company Thyssen-Krupp-Uhde. One of the projects carried out by that company was a study of a 111 MW_{fuel} HTW gasifier for production of methanol by gas from biomass in Sweden.

During the last 30 years many gasification units of the same kind as the Winkler gasifier have been built, employing biomass. However, most of them were stopped for economic reasons. An example of a recent successful plant is the 2×80 MW_{fuel} gasifier in Lahti, Finland [3] using biomass and sorted waste. The product gas is cooled and filtered at about 400 °C before it is burned in a boiler. This arrangement has several advantages: the filter temperature permits removing the fly ashes in a solid form, including alkali compounds and many of the heavy metal contained in waste. At the same time most of the tars do not condense.

Hence, the connected boiler burns cleaned gas and is no longer a waste boiler, which otherwise would have made the boiler more expensive and prevented co-combustion with other types of fuel. Moreover, the corrosion propensity of the gas is reduced and high steam data can be used, which increases the efficiency of electric power production compared to what is normal in waste boilers.

Combustors

Bubbling fluidized bed (BFB) boilers at atmospheric pressure

The first effort to burn solid fuels in a fluidized bed was made in the Soviet Union after World War II [4] with the purpose of developing boilers for industry and district heating. Figure 3(a) shows a drawing of one of the prototypes. The coal was to some extent gasified in the bed and no horizontal cooling tubes were used there; the gases produced were burnt in the freeboard by secondary air.

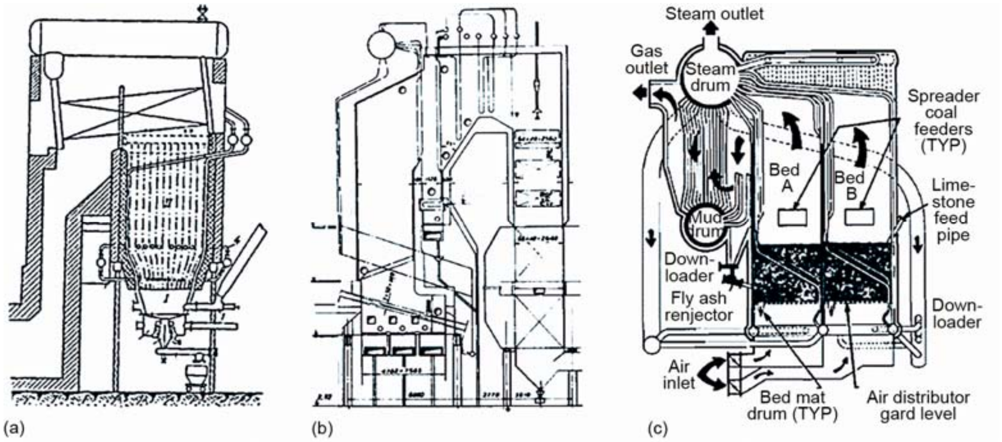

Figure 3. (a) FBC boiler built in the USSR in 1950s [4], (b) Chinese 130 t/h FBC boiler from 1980 [6], (c) One of the most developed boilers in this period, Georgetown FBC 50 t/h boiler, 1980 [7]

There was a problem, probably, to avoid slag formation, because in a later publication [5] a modified design is shown where the fuel is fed on top of the bed, and various ways to control the bed temperature, among them cooling tubes, are discussed. However, before any commercial plant was built, natural gas became more attractive in the Soviet Union, and the FBC development lost its momentum.

Instead, the FBC technology was introduced in China, and thousands of small FBC units were operating there already during the 1980s [6]. Figure 3(b) gives an example. Because of the potential environmental advantages and the ability to burn various types of fuel, the development of BFB combustors (operated at "low" gas velocities, 1-3 m/s) was taken up also in USA and Europe [8]. One of the most advanced BFB designs from this region is shown in fig. 3(c). Further examples from the early activities are found in the book of Oka [9].

After some years of development it became obvious that BFB was a dead end for coal combustion because of several disadvantages: (1) heavy erosion on in-bed heat-transfer

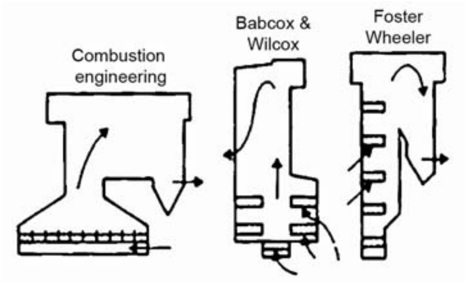

Figure 4. Efforts to design large-scale boilers using BFB [11]; the beds are located in the bottom part or in cages mounted on the walls

surfaces; (2) very large bed surface was needed for utility-size boilers, fig. 4; (3) considerable loss of combustible char; and (4) the sulphur capture was not as good as anticipated.

In a BFB the limestone is contained in the particle phase of the bed, subject to predominantly reducing conditions, while gas containing oxygen and SO_2 passes through the bubbles. In a circulating fluidized bed (CFB) the particle phase, even in the bottom part, is more disperse and tends more towards oxidising conditions, favouring sulphur capture. Particles are also contained in the riser allowing further reactions. As a result, the sulphur capture is more efficient as seen from fig. 5 [10]: the retention is higher at a smaller Ca/S ratio than in a BFB. Because of changes in the reducing/oxidizing conditions the optimum is displaced towards higher temperatures in CFB.

However, the BFB did not disappear: the drawbacks of coal combustion identified above are less important for biomass and organic waste, both high-volatile fuels, which tend to burn and release their heat in the freeboard above the dense bed to a large extent, allowing operation of bubbling beds at reasonable bed temperatures (750-900 °C) without immersed cooling tubes. So, BFB presently burn biomass and waste in boilers of up to a few hundred MW_{th} in industrial and district heating systems [12]. A typical example is given in fig. 6, where the temperature profile reveals how the heat is released in the furnace. Air is added to the bottom bed and to secondary-air nozzles in the freeboard. The bottom air meets the volatile gases and reacts above the bed, while combustion in the bed itself is moderate, maintaining a relatively low bed temperature. The final combustion, promoted by the secondary air, creates a temperature peak, which might lead to melting of ashes, forming deposits on the walls. Therefore, to avoid excess temperatures, the secondary air is often added in stages. The boiler in fig. 6 shows an empty gas pass before the gases enter the convection section. This could have two reasons: (1) if the boiler is to be used for (prepared) wastes, then (in the European Union) there must be a gas residence time of at least 2 seconds after the last supply of air

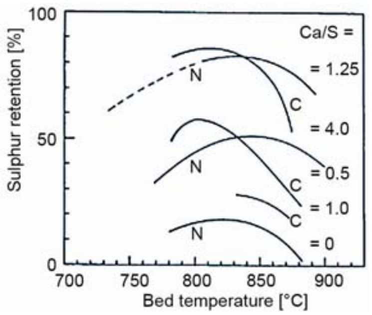

Figure 5. Sulphur capture in a BFB boiler (16 MW_{th}) (C) and a CFB (40 MW_{th}) boiler (N) operated with coal under similar conditions and with the same limestone [10]

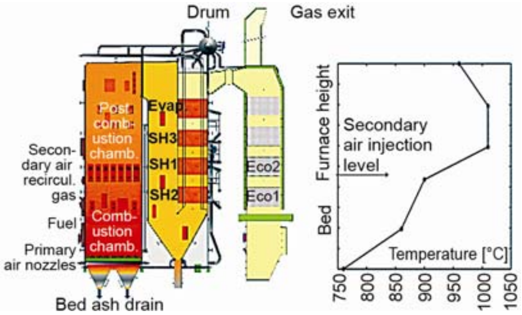

Figure 6. The BFB boiler for biomass and waste, showing the temperature profile in the combustion chamber

at above 850 °C according to EU's Waste Directive [13]; (2) to reduce the propensity for corrosion on the final superheater, the designs often optimize the gas temperature to be relatively close to the tube temperature at the entrance of the superheater tube bundle.

Bubbling fluidized bed boilers at higher pressure (PFBC)

The development of PFBC up till 1992 has been clearly described in [14]. Three large-scale pilot plants were put into operation in 1991 in Spain (Escatron), USA (Tidd), and Sweden (Vartan). The first two plants are stopped, but the Swedish one is still in operation as a combined heat and power plant. The last unit built was the Karita 365 MW_e plant in 2001 in Japan. The experience was relatively positive but some drawbacks became obvious: (1) despite low fluidization velocities some erosion of in-bed tube bundles was experienced; (2) increase of the flue-gas temperature to levels permitted by modern gas turbines requires an additional combustion chamber and perhaps a gasifier to produce the gas needed to be burnt for the enhancement of the flue gas temperature; although this can be done, it is a complication, and furthermore, desulphurization of that gas adds to the complexity; (3) cleaning of the gas by cyclones is possible, but filters are a better solution to protect the gas turbine from particles and some alkali compounds. However, even if considerable development efforts were made, reliable high-temperature filters are still not available. Despite the "low" temperature before the gas turbine in the plants mentioned, efficiencies of 42% were reached. The thermodynamic advantage of PFBC remains, but the road chosen in the recent development of FBC is efficiency enhancement in atmospheric pressure CFB employing supercritical steam data.

Circulating fluidized bed (CFB) boilers

It was found that operation at higher gas velocities (5-6 m/s), returning the entrained bed material to the furnace by a particle separator (CFB), is less susceptible to the problems mentioned in relation to BFB: (1) heat is transferred by the entrained particles to the vertical wall surfaces of the combustion chamber and to additional vertical surfaces inserted in regions less exposed to erosion than in the dense fluidized bottom-bed; (2) higher energy density (MW/m^2 cross-section) makes the combustion chambers narrower, and therefore the scale-up to larger sizes becomes easier; (3) better combustion and sulphur capture efficiencies are attained through the longer residence times for small particles achieved by re-circulation.

A dominant design criterion in FBC, as well as in CO_2 capture processes involving solid fuel, is the large gas volume-flow that passes the equipment, especially at fuel-conversion temperature, 800-900 °C and atmospheric pressure. Just like illustrated by the BFB in fig. 4, the bed cross-section becomes large unless the gas velocity is increased like in a CFB. So, from the mid-1980s CFB became the predominant design for combustion of coal (also for other fuels), employed in a variety of ways, as illustrated in the survey of fig. 1, including conventional CFB for coal combustion as well as various modifications for CO_2 capture. In low-temperature devices for CO_2 removal, applied in the cold end of the flue-gas pass, the situation is slightly relaxed: the volume flow is only one third to one fourth of that at high temperatures (300 K/1200 K), but CFB has been proposed for many applications in this region too.

The first patent on high-velocity fluidization was obtained by Lewis and Gilliland [15], who realized that they could achieve greater output from their catalyst-bed reactor if they increased the fluidization velocity above that normal for a low-velocity fluidized bed. They found that high input of solids to the bed was necessary to maintain a relatively dense particle suspension in a tall riser and to avoid emptying the bed. They did not mention the design of the reactor, but in an accompanying patent Lewis [16] described a circulating system, fig. 7.

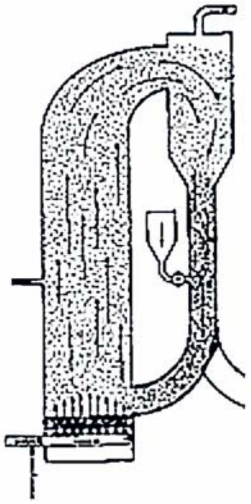

Figure 7. Lewis' CFB proposal [16]

Lewis was the first to propose a concept similar to CFB. Lewis' and Gilliland's inventions were applied in the fluid-bed catalytic cracker of Standard Oil in Baton Rouge 1942 [17], but it took two decades for the first designs reminding of today's CFB for combustion of solid fuels to appear in a patent on a reactor for calcination of aluminum hydroxide by the German company Metallgesellschaft AG [18] (from their subdivision of the name of "Lurgi"). A calciner is a furnace where fuel is burnt to provide heat for calcination and not for steam production like in a boiler, but otherwise the principal features are the same as those of a CFB boiler. So far no combustion application for solid fuels was mentioned; the focus was on calciners, and the first industrial calciner was put into operation in 1970 in Lunen, Germany [19].

During the 1970s several patents were issued, displaying various aspects of CFB combustors, for instance, for waste combustion [20], fig. 8(a). A similar CFB concept was patented by Yerushalmi *et al.* [21], fig. 8(b), which was explicitly said to have a fast bed, referring to relevant scientific literature published by Yerushalmi himself [22] and Reh [23]. Many of these designs separated combustion in the furnace from heat transfer in an additional bed, a BFB (called "slow bed" by Yerushalmi) in connection to the downcomer from the cyclone. Often there is no loop seal on the patent drawings. The most important patents related to boilers belong to Metallgesellschaft [24, 25], fig. 8(c), based on the previous experience of that company on calciners.

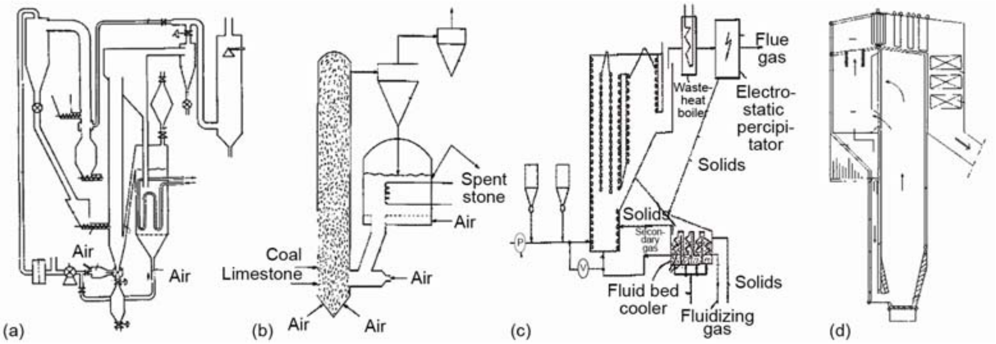

Figure 8. Some early patents on CFB boilers; (a) Stahl, Becuwe 1972-74: Combustion of industrial or household wastes [20]; (b) Yerushalmi, Erlich 1977-78: Combustion of carbonaceous fuels by a fast and a slow bed [21]; (c) Collin, Flink, Reh 1977-79: Process for burning carbonaceous materials [24, 25]; (d) Hyppanen *et al.* 1991-92: Centrifugal separator [27]

The Lurgi patents [24, 25], fig. 8(c), having the priority date of 1975, comprise reactor, separator, recycling conduit for particles, and air supply, organized into two streams: primary and secondary air, such as is common in combustors of all types. The secondary air could be added in several locations along the height of the furnace. The lower part of the bed could be downwardly tapered, *i. e.* with inclined walls. There is an external fluidized-bed heat exchanger attached to the system, but heat could also be transferred to tubes in the furnace and in the particle separator. The combustion bed is characterized by a certain particle density-gradient from the bottom and upwards. It was claimed that there is no jump in the transition in density between the denser phase in the bottom region and the upper dust-containing space in accordance with the characteristics of a "fast" fluidized bed (*i. e.* no distinct bed surface). In fact, the authors mentioned [22, 23] and Kwauk [26] from the Chinese Academy of Science, independently, were the predominant advocates of the fast-bed concept that came to play an important role in the interpretation of the fluidized regime prevalent in a CFB reactor.

After these significant development steps of CFB, many patents were awarded, focusing on details of the process. In fig. 8(d) just one example is given of a subsequent development: the integrated particle separator, which considerably simplified the boiler structure, and contributed to the market success of the company involved, Foster Wheeler (after purchasing Ahlstrom Pyropower in 1995, the company that was originally awarded the patent). This cyclone was made up by water-wall panels just like an extension of the furnace and could have the cross-section of a "square, rectangle, or other polygon". In the first boilers the cyclone was quite square-ish, while subsequently it has been given a more rounded form, still employing refractory-coated tube panels.

Figure 9 illustrates some principal actors in the field of the development of CFB but, indeed, not all: in 1987 Makansi and Schwieger [28] reported on 55 manufacturing firms (excluding Chinese) engaged in the development of FBC, so, obviously, the diagram is simplified. In the centre, the Metallgesellschaft/Lurgi branch is seen. The step was not too far from calciners to building the first coal-fired CFB boiler in Lunen (84 MW_{th}) 1982 followed by a 208 MW_{th} plant in Duisburg (heat and electric power) 1983 [19], which was successfully operated for several decades. The shape of these plants reflects the previous work on calciners: refractory-lined combustion chamber with external heat exchangers for heat recovery. The cyclones in Duisburg

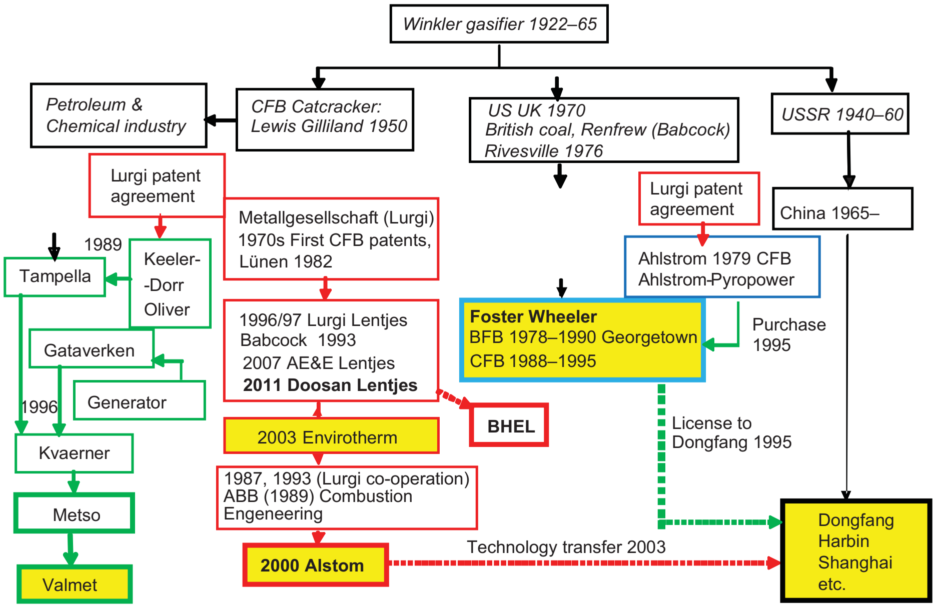

Figure 9. Some CFB manufacturers and their relations (mostly US-EU and those active during the early development)

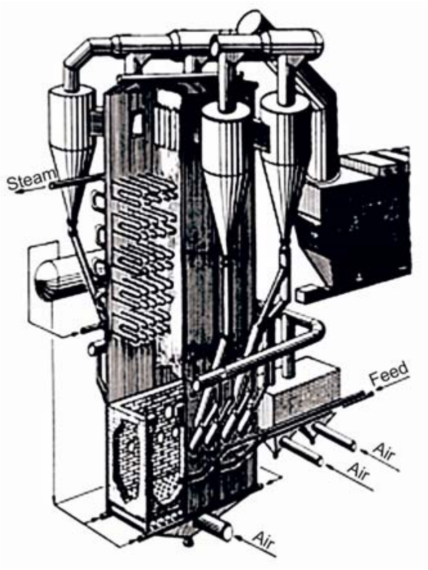

Figure 10. The original Lurgi CFB boiler intended for shale combustion [19]

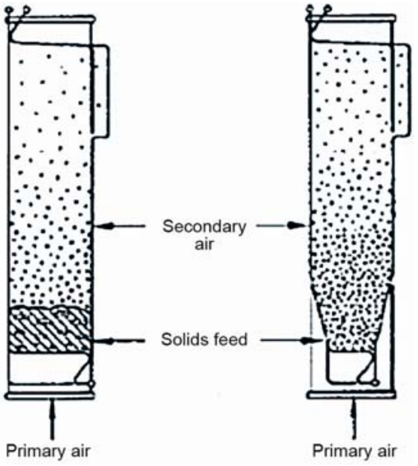

Figure 11. The pronounced CFB according to Foster Wheeler (left) compared to the "fast bed" [34] (right)

were in series of two, one of which proved to be redundant and could be removed very soon after initiation of operation. Later Lurgi joined boiler makers and the design took the form seen today.

Before the Lunen plant was built, Lurgi, together with Swedish co-workers, was developing a boiler for combustion of Swedish oil-shale for extraction of uranium [29, 30]. The corresponding patent, applied for in 1975 [31], showed a sketch similar to that of fig. 8(c) but the actual concept looked more like a boiler, fig 10.

Sketches of CFB boilers were published quite early. In 1977 a simplified image of a "fast" fluidized bed combustor for shale combustion in connection to uranium extraction was shown without comments [29], while a more detailed picture is presented in [30], complemented by several earlier references to CFB. Because of political reasons nuclear energy and extraction of uranium were stopped in Sweden, and the CFB project was never carried out.

The concept and design were developed by Lurgi before Folke Engstrom at Ahlstrom Oy in Finland started experiments in 1976 with the intention to develop a CFB [32]. The work was successful, and in early 1979 a 15 MW_{th} CFB plant was put into operation with bark, wood waste, and coal as alternative fuels. In the following period several boilers with increasing size were built by Ahlstrom Oy. After some time, the company established itself in the USA under the name of Ahlstrom Pyropower and successfully continued to produce CFB boilers until it was purchased by Foster Wheeler in 1995.

Undoubtedly Ahlstrom was the first to build a CFB boiler for combustion of solid fuels. This development was carried out initially without knowledge of the Lurgi patents and without applying for own patents [33]. Inevitably, this resulted in a requirement to establish some kind of patent agreement with Lurgi, allowing Ahlstrom Pyropower to continue building CFB boilers. Until the patents expired in the beginning of the 21st century, Lurgi defended its rights also *vs.* other boiler makers, which were obliged to sign patent agreements in order to use the CFB process. An exception was Foster Wheeler (before 1995) designing a CFB process, which was sufficiently different from that of Lurgi to avoid breaching the patent conditions, the "pronounced bed" process, fig. 11 (left). The differences were explained as follows [34]: "The Foster Wheeler process is characterized by the presence of a pronounced bed in the bottom few feet of the furnace and a relatively solids-lean freeboard above it. An alternative process, *i. e.*, fast fluidized or highly expanded bed, is characterized by having the solids spread over a substantial height of the furnace with the absence of a pronounced bed at the bottom of the furnace …". The "alternative process" coincides with that described in the Lurgi patent. A sketch of such a boiler is shown in fig. 11 (right).

It is interesting to see how ideas can occur spontaneously in parallel. Here is another example.

During the development of catalytic crackers Gilliland (see above), quoted by Squires [35], wrote: "Neither Dr. Lewis nor I have ever seen a Winkler gas producer and it was not the basis of our discussion". Probably, at the time of their invention the concept of fluidization was not in their minds, and they were approaching their problem independently. However, they quoted the Winkler patent in their patent application [15]. This is not a great issue: their invention differed in many respects from that of Winkler, but it was reasonable to mention him in the patent document.

The many subsequent activities in relation to the Lurgi design, indicated in fig. 9, were initiated by the bankruptcy of Metallgesellschaft 1993, which stopped their department Lurgi, although the boiler activity had been successful. A number of firms and constellations followed, which also led to a dispersion of the technology in the form of licenses. The most important company in this chain of transformations is today Alstom. The other presently important manufacturer, independent of Lurgi, Foster Wheeler, got a late start in the development of CFB 1988, but recovered after purchasing Ahlstrom Pyropower in 1995.

To the left on fig. 9, a great number of firms are seen, which converged into Valmet, another Finnish company, after mergers and commercial transformations. The Chinese development, seen on the right-hand side of the diagram, accelerated, especially after the technology transfer agreement with Alstom 2003 that resulted in the 300 MW_e plant in Baima, which was followed by an "explosion" of new large-scale Chinese plants. At present, the largest boiler in the world, 600 MW_e, designed by Dongfang [36], has been taken into operation in China. The Chinese development, including Chinese research related to boilers, has recently been described by Yue [37].

Resulting CFB designs

The predominant designs of CFB boilers, after gradual development, converge into rather common features, irrespective of manufacturer: (1) the size of the air distributor at the furnace bottom is about half of the cross-section of the upper part, an effect of the inclined front and rear walls. About half of the air is added through the bottom and the rest at one or two levels a few meters above the bottom. The fuel is added in various points along the tapered walls and sometimes also in the return ducts of bed material from the cyclones with one feed point per 20 to 30 m^2 furnace cross-sectional area for large boilers (To be compared with one feed point per m^2 which was the recommendation for coal-fired BFB). The arrangement with tapered walls reduces the consumption of electric power compared to a design having straight vertical walls. It also improves mixing of fuel and air across the cross-section of the furnace; (2) The width of the furnace is limited to less than 10 m by the penetration of the secondary-air jets and the spread of the fuel, clearly seen in larger boilers where the furnace cross-section is given a rectangular shape; (3) A cyclone is the most efficient form of particle separator, and CFB boilers have cyclones for particle re-circulation, with a few notable exceptions, for instance Babcock&Wilcox' U-beam separator [38]. The cyclone is a centrifugal separator and its efficiency is inversely proportional to its radius. Therefore, and for space

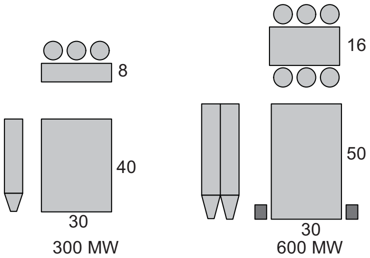

Figure 12. Scale-up from a 300 MW$_e$ boiler with three cyclones (left) to a hypothetical 600 MW$_e$ plant (right), formed by the addition of two 300 MW$_e$ furnaces

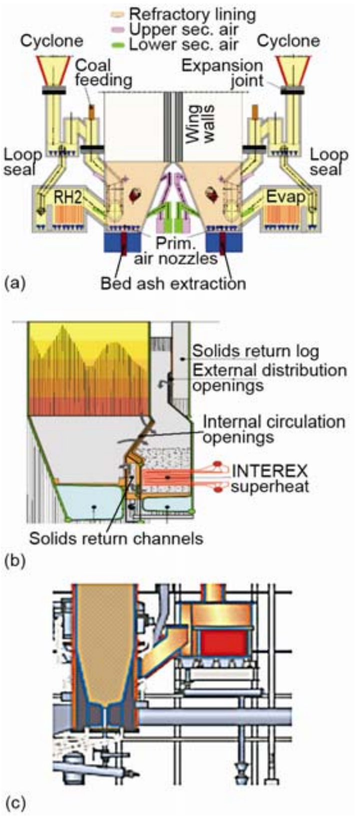

Figure 13. Examples of external or loop seal heat exchangers from various manufacturers; (a) Alstom; (b) Foster Wheeler, (c) Valmet

reasons, large CFB boilers have several cyclones. There are two options: cold or hot cyclones. The cold cyclones are connected to the steam-water system of the boiler and are steam or water cooled. The hot cyclones are simply refractory structures connected to the furnace through thermal expansion compensators; (4) Internal heat transfer surfaces are wing walls located in the upper part of the furnace or internal walls extending from bottom to top. In very large boilers external heat exchangers of some kind may be used; (5) In the largest boilers built recently, the steam data are increased to the supercritical range, important for the efficiency of electricity production but not significantly affecting the combustion side of the boiler.

Scale-up in size is an important trend during recent years. Originally, the membrane-tube walls of the furnace served as the only heat-transfer surface, but during scaling up to larger size, to more than 50 MW$_{th}$, the wall surface area is insufficient, despite the fact that for large boilers the furnace is made as tall as is considered reasonable, a maximum height around 50 m, fig. 12. Above that height, the particle concentration has declined too much, and it may be difficult to maintain the bed temperature and the heat transfer in the uppermost parts. To extend the heat-transfer area, internal surfaces (wing walls, division walls, *etc.*) are introduced, located in parallel to the particle flow. Finally, in large plant the return flow of hot particles from the separator is used for heat transfer, as shown in fig. 13, either through heat exchangers included in the loop seal, cf. figs. 8(a) and 8(b), or in external heat exchangers, cf. fig. 8(c). Figure 13 indicates that designers prefer to use the more protected region in the return loop, even though the capacity of a loop-seal heat-exchanger is limited. In fig. 12 (right) it is shown how a 600 MW boiler could be formed from two 300 MW boilers. Among several minor adjustments needed, the more limited space for wing walls in the large furnace, surrounded by cyclones, has to be compensated by additional internal vertical heat-transfer panels and external heat exchangers, in separate arrangement like in fig. 13(a) or in loop seals like in fig. 13(b) or (c). The common wall may be replaced by a dedicated internal tube-wall.

Development trends

Just like FBC was once introduced to the Western countries because of its anticipated advantages with respect to low emissions of pollutants, the recent development trends utilize the advantages of fluidized bed for devices aiming at CO_2 reduction. These new development trends can be divided into single-reactor systems (oxy-fuel combustion) and dual-reactor systems (chemical-looping combustion, calcium looping, and several other systems consisting of two reactors). Dual reactor systems have been used in the petroleum industry for a century. For gasification of biomass and waste it was first introduced in the beginning of the 1970s in Kunii's Pyrox plant [39] (see fig. 1), consisting of two coupled fluidized beds. In the recent developments, this principle is employed both for coal and for biomass gasification pilot plant. Below, some proposals for CO_2 reduction and biomass gasification will be described briefly. So far no large-scale plant for CO_2 reduction has been built.

Oxy-fuel combustion

Conventional CFB designs fed with oxygen, single FB system

An oxy-fuel CFB is a regular FBC boiler supplied with pure oxygen instead of air to produce CO_2 in the flue gas, not being diluted by nitrogen. To replace the nitrogen and to moderate the O_2 concentration, a certain amount of the flue gas is re-circulated either as dry gas (CO_2) or as wet gas ($CO_2 + H_2O$), as seen in fig. 14.

The application for CO_2 capture may have two forms: (1) the "CO_2-capture-ready" concept, which is similar to present air-fired CFB boilers prepared for CO_2 capture, if demanded; (2) "new design" where the oxygen concentration is chosen as high as possible in a new boiler. The CO_2-capture-ready design replaces N_2 with CO_2, maintaining the design of the air-fired boiler with the same bed and entrance temperatures T_{bed} and T_{in}. Also excess oxygen and fluidization velocity are kept unchanged at given power and furnace dimensions. To fulfill the heat balance, the gas flow g times the specific heat c_p has to be identical in the air-fired (af) and CO_2 circulation (CO_2) cases:

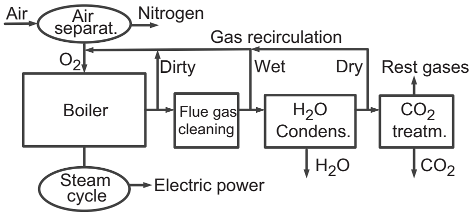

Figure 14. Oxygen supply to a CFB furnace from an air separation unit; the flue gas can be cleaned and dried before re-circulation

$$(T_{bed} - T_{in})(gc_{pgas})_{af} = (T_{bed} - T_{in})(gc_{pgas})_{CO_2} \tag{1}$$

In the dry circulation case $c_{paf} = 1.098$ and $c_{pCO_2} = 1.090$ kJ/kgK, that is, they are almost equal. The gas flows in the air and CO_2 circulation cases should then also be about equal, $g_{af} \approx g_{CO_2}$, which is achieved by the re-circulation of flue gas. To maintain the heat balance in the air-fired case a mass concentration of oxygen of around 30% in the inlet gas is required, somewhat depending on the excess oxygen chosen [40]. In order to keep the oxygen excess in the furnace similar to that of the air-fired case, the excess oxygen supplied can be smaller than in the air case [40], because the circulated oxygen helps maintaining a desired level in the furnace.

In contrast to an oxy-fired pulverized fuel boiler, in a CFB of the CO_2-ready case there is an additional criterion to be fulfilled: the fluidization velocity should be maintained constant. This, however, is related to the volume flow and contradicts the heat balance criterion eq. (1), which is based on mass flow: both cannot be fulfilled. The impact is not very large

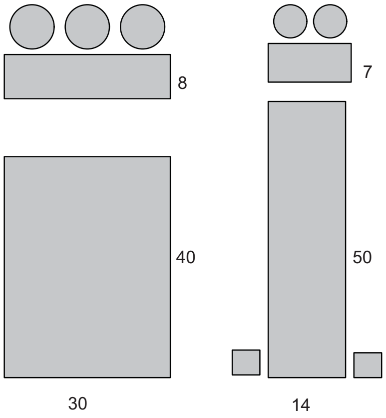

Figure 15. Comparison between air (left) and oxy-fired (right) 300 MW_e CFB at 60% O_2; wall surfaces 240 m^2 *vs.* 96 m^2; the dimensions are taken from a typical air-fired 300 MW_e CFB boiler and the height of the oxy-fuel boiler is extended as much as reasonable [40]

and the remedy is to relax somewhat the strict equalities between temperatures and oxygen supply declared above.

The new-design case is straight-forward: for a given power most parameters are chosen similar to conventional data except the excess oxygen, which can be lower than in air combustion for the reason just mentioned. The amount of gas re-circulated at a given bed temperature and fluidization velocity depends on the chosen oxygen concentration in the input gas. The higher the oxygen concentration, the less nitrogen has to be replaced by flue-gas re-circulation: the total gas flow will be smaller, and to attain the given fluidization velocity the cross-section of the furnace should be made smaller, as seen in fig. 15 where an oxygen concentration of 60% gives half the furnace volume of a corresponding air-fired boiler based on gross power (power consumption for air separation and CO_2 compression for disposal have not been included). There will be less space for heat transfer surfaces, and external heat exchangers may be necessary.

The decisive factor, so far not well known, is the rate of particle circulation to transport the particles (heat) to the external heat exchangers. In the literature there are few data on the rate of particle circulation in boilers, and those available are quite divergent, ranging from 5-10 [41] to 30-40 [42] for boilers of the kind treated here.

Chemical looping combustion (CLC), dual FB system

In CLC, the drawback of the above type of oxy-fuel CFB, the air-separation unit, is absent. Instead, metal particles are oxidized in an air reactor (a CFB unit) and transported to another FB reactor where the particles are reduced by reacting the fuel. This oxy-fuel boiler, seen in fig. 16, belongs to the dual-reactor group consisting of an "air reactor" where oxidation takes place and a "fuel reactor" where the oxygen brought by the oxidized particles is consumed by the fuel. The present status of this method has been thoroughly reviewed [43, 44].

The CO_2 in the off-gas from the "fuel reactor" is free from nitrogen because the oxidation of the metal powder in the air reactor by air leaves the nitrogen in the flue gases from that reactor. The metal, now in the form of an oxide, is transported to the fuel reactor where it is reduced by the fuel, which "burns", yielding the gaseous combustion products H_2O and CO_2 in the off-gas to be treated for CO_2 deposition. The ashes are removed, and the metal is returned to the air reactor for renewed oxidation. With most oxygen carriers, the fuel energy is released during the oxidation, and the air reactor serves as a boiler absorbing reaction energy and providing sensible heat to the fuel reactor by the oxygen carriers. The total heat release is the same as in conventional combustion.

There are three major paths of reaction in the fuel reactor, as illustrated in fig. 17. The application with gas as a fuel is simpler than with solid fuels. The fuel gas, serves as a fluidization medium, and reacts directly with the metal oxide. The stoichiometric air-fuel volume ratio indicates that the air volume is much larger than the fuel volume, and the CFB air-reactor could be coupled to a BFB fuel-reactor. The conversion of solid fuels (particularly

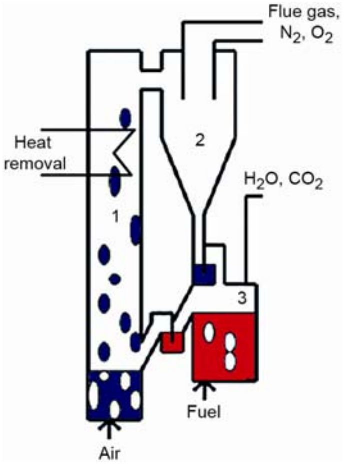

Figure 16. CLC reactor system for gaseous fuel; (1) air reactor, (2) cyclone, (3) fuel reactor, (4) loop seals. In designs with solid fuel the fuel reactor is a CFB fluidized by added steam or by steam/CO_2 from the off gas. Modified from [46]

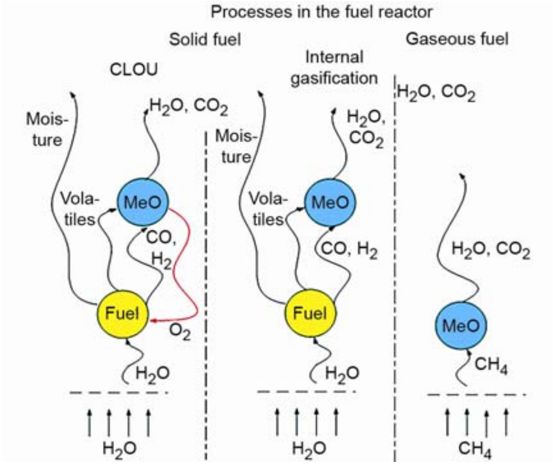

Figure 17. Three cases of reaction in the fuel reactor. (right) A gaseous fuel reacts with a metal oxide particle. (center) A solid fuel particle is gasified and then the gases react with the metal oxide particle (MeO). (left) A metal oxide particle releases oxygen and enhances thereby the reaction. Modified from [46]

the char part) is more complicated. Solid-solid reactions are out of question. So, the char has to be gasified by steam and/or CO_2, which also serve as the fluidization medium. Gasification is a slow process at the temperature concerned, but enhancement is possible by the release of oxygen from some types of oxygen carrier for thermodynamic reasons, while moving from the oxidizing conditions in the air reactor to the reducing conditions in the fuel reactor. The oxygen released reacts readily with the fuel. Such a process is called CLOU [47]. Other enhancement of a catalytic nature has been observed for some oxygen carriers [48]. Furthermore, the rate of gasification tends not to be inhibited by H_2 and CO as usual in gasifiers, since these gases are consumed by the oxygen carriers.

Preferably also the fuel reactor should be a circulating bed, because the fuel's volatiles in particular tend to react above the surface region of a BFB, and the contact with the oxygen carriers is limited while the combustible gases ascend in the freeboard. In the circulating bed the oxygen carriers are present in the entire reactor, although it is desired that the solids density be enhanced in the upper part of the riser [49].

There are a number of complications that have to be solved by additional measures. The most important ones are: (1) The ashes of solid fuels have to be separated from the oxygen carriers. Such a separation process is not perfect, and therefore only cheap oxygen carriers, which can be wasted to some extent, should be used; (2) In any case, the bed material (the oxygen carrier) contains ashes and unburnt char, which has to be prevented from returning together with the metal particles into the air reactor. Some burn-out device might be needed in between the two reactors; (3) Unburnt gases and some char may escape with the CO_2 from the fuel reactor, but in a CFB this amount can probably be reduced to a low level.

On the whole, CLC is a promising technology, if the complications mentioned do not seriously impede the process, and high combustion efficiency can be attained also with solid fuel. Although it has not yet reached the pilot plant scale, one can imagine how a power plant using this technology would look like: in [50] a tentative design and cost analysis is carried out.

The CO_2 removal from flue gases (post combustion CO_2 capture), dual FB systems

High-temperature systems (Calcium looping)

Calcium looping is a post-combustion CO_2 capture process consisting of two coupled fluidized-bed reactors, as suggested by Shimizu *et al.* [51]. Overviews of this method have been presented in [52, 53]. The CO_2 contained in the flue gas from any type of boiler is absorbed by CaO converted to $CaCO_3$ in a CFB reactor (the "carbonator"), fluidized by the same flue gas. The CaO needed is produced from added limestone, $CaCO_3$, and recycled material from the carbonator to another FB reactor (the "calciner") while CO_2 is released for deposition. From there the CaO produced is returned again to the carbonator. The corresponding reactions take place when temperature and pressure in each reactor are within the ranges of the respective chemical equilibria, as shown on fig. 18.

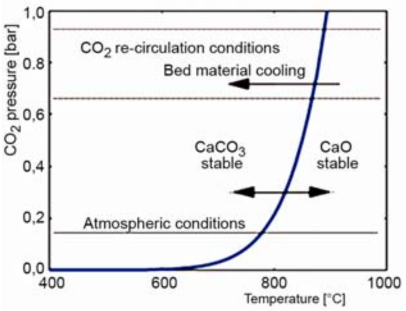

Figure 18. Equilibrium regions of calcination and carbonation in respective reactor, shown as CO_2 concentration *vs.* temperature

This flue-gas treatment method appears attractive because limestone is readily available in many locations, and presently a great deal of research has been carried out to analyze this process, which is an alternative to amine scrubbing to remove CO_2 from flue gas. However, there are several severe limitations: (1) the absorption capacity of limestone decays rapidly after some cycles, although this can be compensated for by allowing a sufficiently high circulation rate [54] and addition of make-up limestone; (2) the size of the equipment is large. Flue gases at 650 °C, which is a typical carbonator temperature, have a considerable volume. Strole *et al.* [55] estimated the cross-section of the carbonator to be about 400 m^2 for a 1000 MW_e power plant, despite rather favorable assumptions (45.6% net efficiency of power production and 6 m/s fluidization velocity); (3) the calcination reaction is endothermal (178 kJ/kmol) and heat has to be supplied to the calciner by combustion, which converts the calciner into a coal-fired oxy-fuel combustor (but not a to boiler, because the heat produced is absorbed by the reactions), operating at a bed temperature of 900 °C. An air-separation unit is required together with CO_2 re-circulation, just as has been described above for the oxy-fuel CFB process and it is necessary to increase the plant capacity by 50% of the original power plant [55]; (4) char contained in the bed material is transported back into the carbonator with the re-circulating bed material, which reduces the efficiency of the CO_2 capture and may cause incomplete combustion.

In order to appreciate the disadvantages mentioned, one has to compare with alternative post-combustion solutions, such as amine scrubbing by liquid amines. That method also requires equipment of huge dimensions, and it has a considerable energy demand. The essential comparative advantage of calcium looping is that the energy released from the exothermic carbonation is at sufficiently high temperature level to be integrated in the power cycle, whereas the energy from amine scrubbing is released at a low temperature level, making it difficult to find a use for this heat. CO_2 avoidance costs for calcium looping are estimated as lower than those of amine scrubbing [55].

Low-temperature systems

Dry alkali systems. In this proposed process, an alkali compound M (potassium or sodium) reacts in a carbonizer with the CO_2 of the wet flue gas from a boiler according to:

$$M_2CO_3 + H_2O + CO_2 = 2MHCO_3 \tag{2}$$

The bicarbonate produced can be regenerated in a regeneration reactor, fig. 19:

$$2MHCO_3 = M_2CO_3 + CO_2 + H_2O \tag{3}$$

The M_2CO_3 is then re-circulated back to the carbonizer. A description has been given in [56]. There are some similarities with calcium looping, but here the process goes between an exothermic reaction below 100 °C, but above the water dew-point, and an endothermic decarbonization reaction at a temperature of 120 °C to 200 °C higher. The temperature level of the flue gas yields a gas volume, which is about half of that of the calcium looping process, but the heat involved in the process is at low temperature where excess heat is difficult to use.

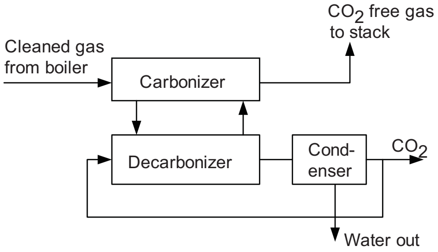

Figure 19. Layout of an alkali CO_2 capture system in the cold flue-gas end of a power plant

Amine capture of CO_2. To improve CO_2 capture by amines, sorbents are prepared by impregnation of solid carrier-particles with amines. These particles are used in a CO_2 removal reactor exposed to the cold-end flue gases and then regenerated in a parallel reactor similar to the previously described arrangement of calcium and alkali looping. An example of a publication dealing with the topic is [57].

Indirect gasification of biomass in dual FB systems

The gasification application with two reactors is called "indirect" in contrast to the Winkler type of "direct" gasification. The heat necessary for gasification is produced by separate combustion in one of the two reactors (the combustor) and transported with the bed material to the other reactor (the gasifier). The flue gas is emitted from the combustor and will not dilute the product gas from the gasifier, which then attains a high heating value without using pure oxygen as would be necessary for the corresponding purpose in a direct gasifier. Biomass and many wastes consist mostly of volatiles, and in such cases pyrolysis of the fuel is a sufficient process in the gasification reactor, while the char is needed in the combustion reactor to produce heat. Imbalance between available char and the fuel to be burnt may require enhancement of gasification or additional fuel feed to the combustor. The reasonably high heating value of the product gas (no nitrogen) is an advantage, while the method shares its disadvantage with most other biomass and waste gasifiers: production of tars requires cleaning, depending on the intended use of the product gas. The Gussing gasifier in Austria [58] is an example of this type, which has succeeded to be introduced commercially in several places, for instance, in the 20 MW_{fuel} demonstration plant in Gothenburg [59], intended for methane production from biomass.

Conclusions

The FBC is now a mature technology, but it is still greatly based on empirical knowledge, and new applications: scale-up, load following, and increased efficiency would benefit from additional and improved basic knowledge.

The CO_2 removal is the most important challenge for further development. Several routes are proposed using FB both in single and dual reactor systems: oxy-combustion, chemical looping combustion, calcium cycling, alkali cycling, and amine absorption with solid sorbents. The research is still on the laboratory scale. Pilot scale plants have not yet been built, but would greatly accelerate development.

Fluidized bed for conversion of biomass and wastes is being gradually improved. Combustion is established, while gasification is fighting with economic obstacles.

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