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Design and manufacturing of a tubular solid oxide fuel cell combustion system

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Abstract

Solid oxide fuel cell (SOFC) stacks are electrochemical, electrical and thermal devices as well. Today there are different developments of planar and tubular SOFC stacks including their integration in power systems. Any successful stack design must yield to a high efficiency, a good thermal performance, a sufficient power density at market prices for the total system and a good heat management including the integration in combined cycles. A new design of a tubular SOFC combustion system is presented and the actually evaluated possibilities of manufacturing are discussed. © 1998 Elsevier Science S.A.

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1. Introduction

A large number of solid oxide fuel cell (SOFC) developments are based on the planar design. There are only some tubular designs under development. The developers of the planar designs claim a lower manufacturing cost of the cells, a lower electric resistance and a higher power density. The main benefits of the tubular design – as claimed by their developers – are the better thermal performance of the cells and the short sealing length of the stack. It is useful to formulate the design requirements as a base of any comparison of the different types of design. Fig. 1 shows the main influences. The design requirements are mainly influenced by the process, the structure to be designed and the manufacturing. A high efficiency of the SOFC stack can be reached by cascades of the single SOFCs. A parallel connected network of SOFCs is the base of a high availability of the stack. The stack must be easily integrated in the system [1] to reach high plant efficiencies and the depleted fuel must be burned at the end of the stack by an integrated combustor. The design should allow a free expansion of the stack components at the thermal cycles during start up and shut down operation to reduce the complicated and expensive use of matched materials. The length of the sealing should be as short as possible and the stack should have a high power density. The cost of the manufacturing process can be reduced by structures and components that can be easily assembled and handled.

2. Design analysis

The design requirements of a high power density of the stack can be seen as a geometric problem in a first order solution. Fig. 2 shows the principles. Any stack can be described as an arrangement of channels of a process flow and a surrounding volume to operate the process flow in the electrochemical device. We assume that the combustion air is the process flow. Any air channel is surrounded by a cylinder with the base area A_{Design}, being necessary to arrange the necessary volume of fuel channels, contacts, separator plates etc., and the length LC of the SOFC as the height. The surface of the air channel consists of an active and a non-active part. If we assume in the first order, that the voltage U and the current density i is independent of the geometric structure of the cell, we consider that the power related volume v_{el}, the reciprocal value of the power density, is only influenced by the ratio V/A_a with the volume V and the active surface or active area A_a.

The single SOFCs are connected serially and in parallel to achieve a sufficient high voltage and to assure a sufficient availability of the stack as well known from the connection of electric batteries. Such a stack arrangement forming a network of single SOFCs is shown in Fig. 3. The fuel flow along the single SOFCs has the same direction as the

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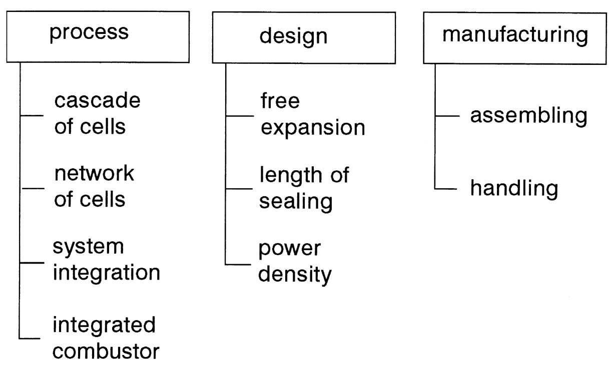

Fig. 1. The design requirements of SOFC stacks.

current flow. The thermodynamic potential of work of the combustion reaction is represented by the Nernst voltage. Due to the increase of the mixing entropy the Nernst voltage of the depleting fuel decreases with an increasing fuel utilization. The voltage of a single SOFC contacted on its surface is, thus, clearly lower than the average Nernst voltage if the SOFC operates with a high fuel utilization. But the Nernst voltage can be approximated by a serial connection of the single SOFCs in a cascade in the flow direction of the fuel. The total voltage of this cascaded SOFC stack is higher and the efficiency increases compared with a non cascaded combustion in a SOFC. Fig. 4 shows the increase of the efficiency as a function of the number of the cascaded SOFCs. The efficiency increase by a cascade increases with the increase of the fuel utilization. The strongest increase of the efficiency occurs in the first three SOFCs of the cascade.

3. Geometric influences

The power density of a planar stack depends on the height Δh as the cumulated thickness of the separator plate and of the planar SOFC, see Fig. 5. The ratio V/A_a of the volume V and the active area A_a – representing the power related volume v_{el} – is defined by the height Δh and the efficiency η_A describing the influence of the covering of the active

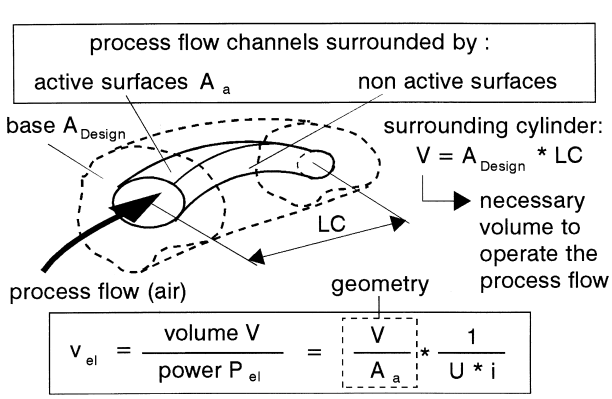

Fig. 2. The principles of the stack geometry.

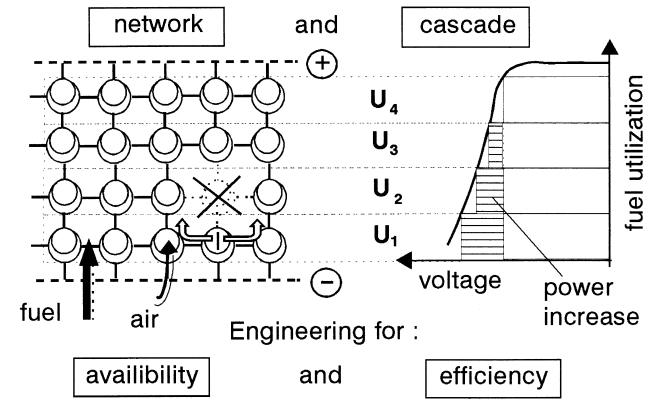

Fig. 3. An example of a SOFC-network with an integrated cascade.

SOFC area A_a by the contacts of the separator plate. We use again the air flow as the reference flow. Fig. 6 shows the results of a variation of the height Δh and the efficiency η_A. For an average thickness of the separator plate and the SOFC of $\Delta h = 4$ mm we get a ratio V/A_a of about 4.

The arrangement of tubes with the highest power density or the lowest power related volume v_{el} is a staggered spacing, where the three neighboring tubes form isosceles triangles. The surrounding volume V of each tube – including the air flow and the fuel flow – is then based on a regular hexagon, as shown in Fig. 7. Thus, the surrounding volume V of a tubular SOFC is defined by the outer radius r of the SOFC tube, by the distance $2s$ between two neighboring SOFC tubes and by the length LC of the SOFC tube. The active area A_a is the surface of the SOFC tube with the outer radius r, see Fig. 7. Again, the power related volume v_{el} depends on the geometric relations represented by the ratio V/A_a. Fig. 8 shows the results of a variation of the SOFC tube diameter $2r$ and the distance s as a parameter. For ratios V/A_a of about 4 we get outer SOFC tube diameters between about 6 and 10 mm, depending on the distance s. In this example the planar design leads to lower values of the power related volume v_{el} or higher power densities for all diameters larger than these values and the tubular design leads to lower values of the power related volume v_{el} or higher power densities for all diameters smaller than these values. However the assumption of a constant $U{*}i$ for a

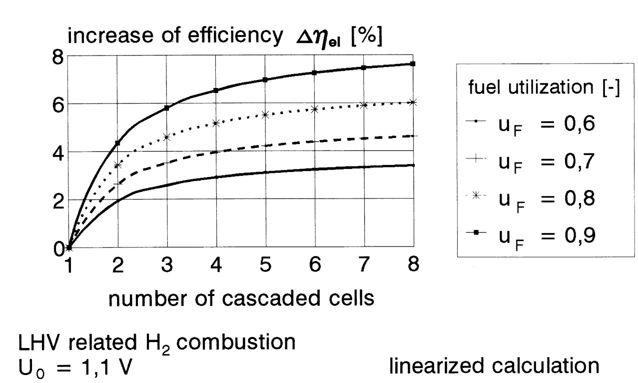

Fig. 4. The efficiency increase by the use of cascades.

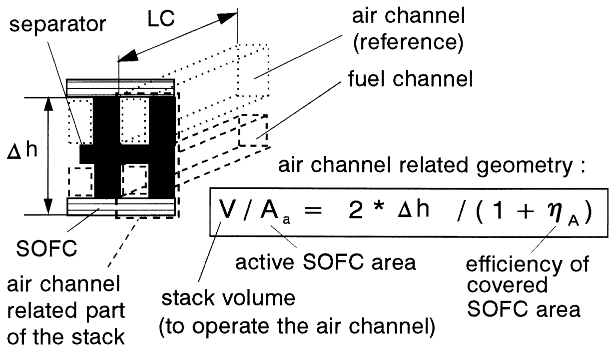

Fig. 5. The determining geometric data of a planar SOFC stack.

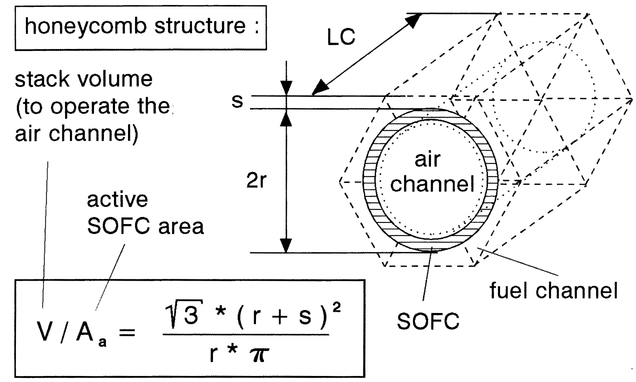

Fig. 7. The determining geometric datas of a tubular SOFC stack.

planar SOFC and a tubular SOFC is only valid for sufficient thin tubes this simple calculation shows that the planar design is not generally the concept with the highest power density.

The length of the necessary sealing is another important design parameter of the SOFC stacks. It is useful for the comparison of the different concepts to define a specific sealing length *SL* as the ratio of the sealed periphery *P* and the active area A_a. The sealing length of a square planar SOFC is defined by the periphery $4*LC_P$ of the square with the side length LC_P and the active area A_a is LC_P^2. The sealing length of a tubular SOFC is the periphery $d*\pi$ and the active area A_a of a tube with the length LC_T is $d*\pi*LC_T$. Fig. 9 shows the context. The specific sealing length *SL* depends on $1/L$ with the characteristic length *L*. The characteristic length *L* for a square planar SOFC is the side length LC_P and for a tubular SOFC the length LC_T of the tube. Fig. 10 shows the influence of the characteristic length *L* on the specific sealing length *SL*. The specific sealing length *SL* of a square planar SOFC is exactly four times larger than the specific sealing length *SL* of a tubular SOFC for a certain *L*.

4. Proposed tubular design

The most common actual tubular SOFC designs are the Westinghouse design [2] and the Mitsubishi design [3]. The Mitsubishi design consists of cascaded single SOFC elements that form the SOFC tube. A parallel connection of the single SOFC elements at the same voltage level and, thus, the forming of a network is not possible. The Westinghouse design can fulfill all requirements to form a network of SOFCs including a cascade by the common connection of the single SOFCs by the nickel felt. However, this design is very successful and further improvements of the tubular SOFC stacks seem to be possible.

The essential element of the proposed stack design is a number of parallel connected SOFC tube elements connected by a nickel felt structure to form a certain voltage level, see Fig. 11. The outer side of the tube is the anode and the inner side forms the cathode. The nickel felt contacts the complete anode surface of the SOFC tube element. The cathode sides of the SOFC tube elements of each voltage level are connected with the anode sides of the SOFC tube elements of the next voltage level by interconnectors to form a cascaded SOFC tube. The nickel felts of the anode side of any voltage level are insulated by an isolating nozzle plate to avoid short circuits. The length of the SOFC tube elements is restricted by the increasing ohmic resistance of the cathode. Calculations show that the length should not exceed about 20–25 mm [4]. More general information about, such calculations can be found in [5]. The use of a thick cathode was also proposed for a planar design to avoid the contacting problems on the air side [6].

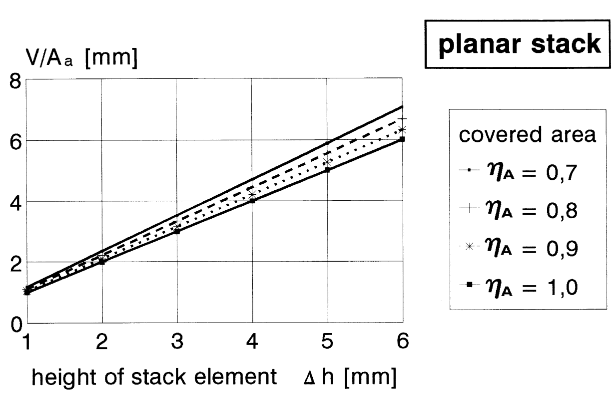

Fig. 6. The ratio V/A_a as a function of the height Δh of a planar SOFC stack.

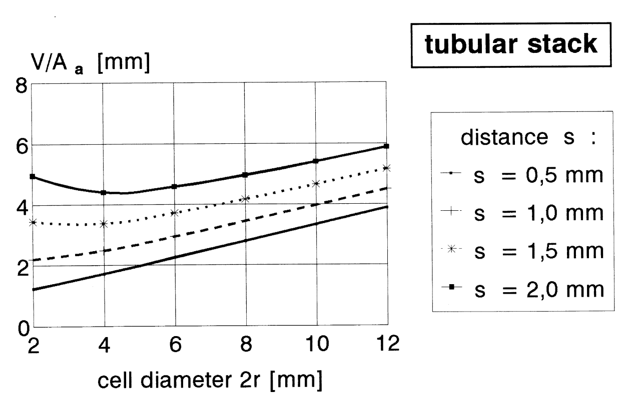

Fig. 8. The ratio V/A_a as a function of the tube diameter $2r$ of a tubular SOFC stack.

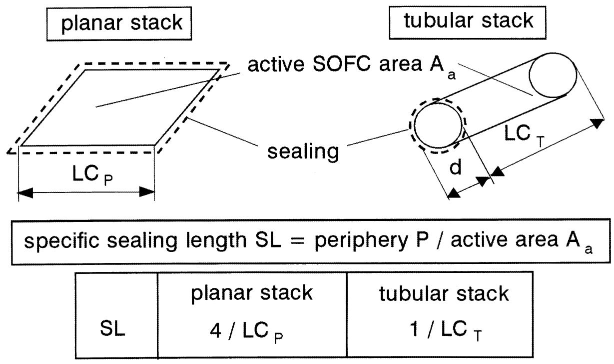

Fig. 9. The principle sealing geometry of SOFC stacks.

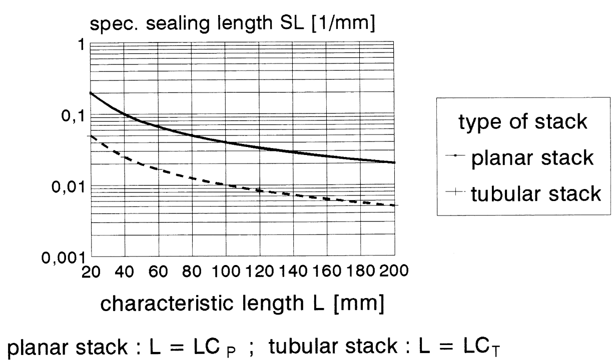

Fig. 10. The specific sealing length *SL* of a planar and a tubular SOFC stack.

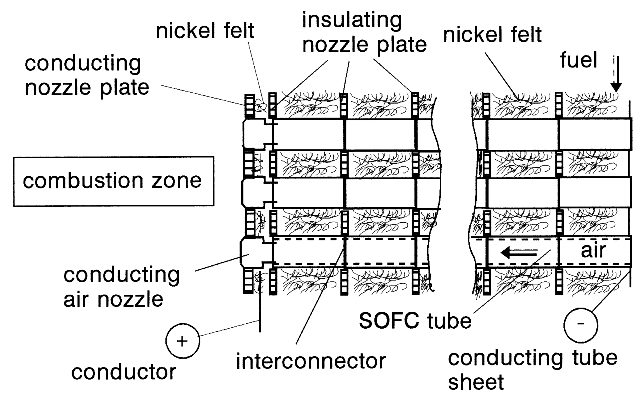

Fig. 12. The proposed tubular SOFC stack with a cascaded network.

The single SOFC tubes are fixed in the conducting tube sheet. The SOFC tubes, the contacting nickel felt and the isolating nozzle plates are assembled to form the stack, as shown in Fig. 12. It is important that the fuel always flood the nickel felt to avoid corrosion. At the end of the SOFC tubes the outlet contacts of the cathode are formed by the outlet nozzles of the depleted air and the surrounding nickel felt. The depleted fuel and the depleted air are burnt in the downstream combustion zone. The design of the stack must be sufficiently gas tight and the structure must be able to tolerate the thermal cycles of the usual load changing conditions of power plants, including the start-up and shut-down operation in a sufficient time. The tubes are only fixed in the nozzle plate at the air plenum. Therefore, a longitudinal free expansion of the tubes is possible. The thermal expansion of the nozzle plates can be matched with the tube expansion. The lateral bearing of the SOFC tubes in the nickel felt is comparably soft.

The integration of these tubular stacks in a power plant system and the arrangement of the integrated heat exchangers is comparable easy as one example shows in Fig. 13. The stack burners can be easily arranged between the pipes of the stack cooling. The cooling pipes can be used for different purposes as, e.g. the fuel preparation or the reheating of the flue gas flow before its expansion in a gas turbine

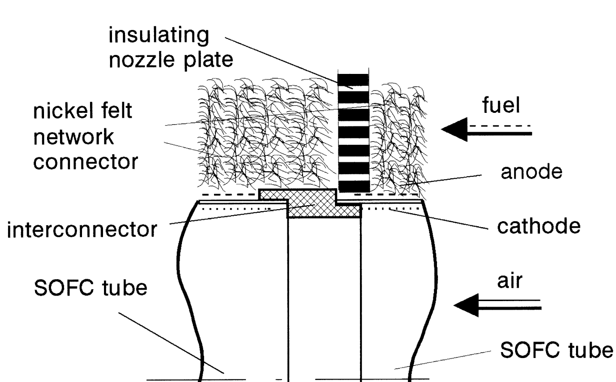

Fig. 11. The basic tubular SOFC element for forming cascaded networks.

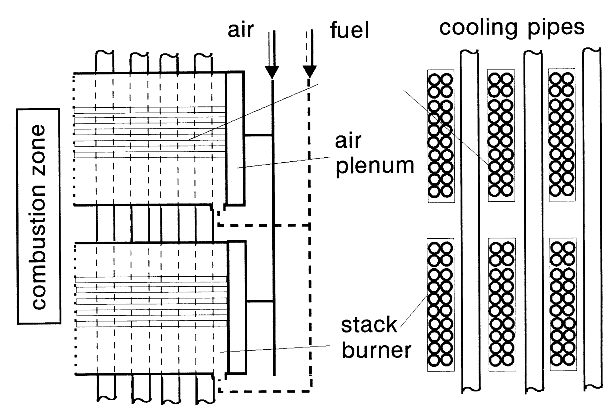

Fig. 13. An example of the process integration of the proposed tubular SOFC stack.

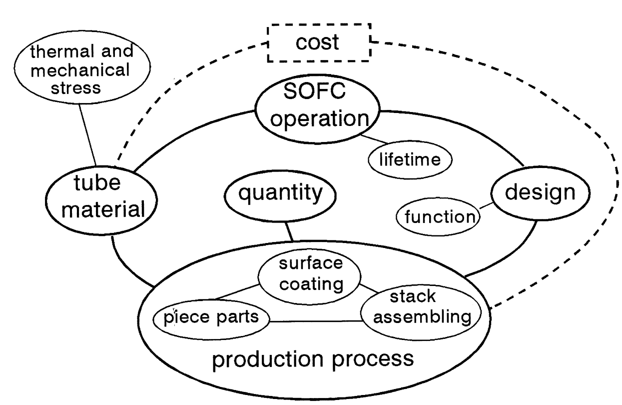

Fig. 14. The interdependencies during the manufacturing process.

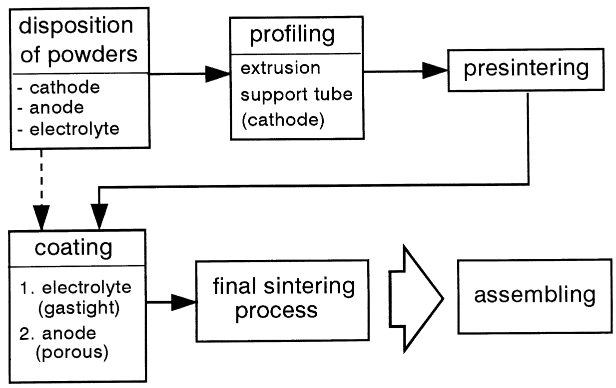

Fig. 15. A possible SOFC tube production process.

[1]. The high flexibility of the design gives the option on a wide variation of the stack arrangement and the stack cooling.

5. Manufacturing of the design

The development of a new SOFC system is mainly influenced by the interactions of the later operation, the different possible design solutions, the different materials and the actual available and commercial production technologies, as the design of any new system. Fig. 14 shows the correlations of this process. This interactive process is finally governed by the cost demands. The quantity of the produced elements is a very important cost factor that strongly influences the production process and the final design as well. The results of the material research support this process with advanced materials and new production processes. This makes clear that a good cooperation between all of these disciplines is a very important factor for the later success.

All parts of the proposed design have a simple geometry to minimize the production cost. The SOFC tube elements and the SOFC tubes are completely centro-symmetric. This has the benefit of comparable simple and cheap steps in manufacturing because the handling and assembling of rotary parts are common and cheap technologies. The key production process is the manufacturing of the tubular SOFC elements. Fig. 15 shows an example of a possible tube manufacturing process. The tube manufacturing is a typical mass production process. The process starts with the disposition of the powders for the production of the cathode, the anode and the electrolyte. The cathode is used as the support tube of the SOFC tube element. An adequate mass production process is the use of an extruder. The extruded cathode tube will be presintered. The tubes used in the first tests will be produced by an isostatic press process, extruding processes will be investigated later. The next step in the tube manufacturing is the coating process to apply the electrolyte layer and the anode layer. The common coating processes for producing the electrolyte layer and the anode layer are, e.g. thermal spraying or wet processes and will be tested. The final sintering process delivers the tubular SOFC tube element. The assembling of the stack is the last step of this planned production line. Our own experience in the design and simulation of production plants will be already used in this early stage of the design and development phase to investigate only economical relevant process chains.

6. Conclusions

The proposed tubular SOFC design has the benefit of forming a cascading of the fuel cell combustion and a SOFC element network in a very simple way. Different ways of system integration of the stacks are possible. The combustion of the depleted fuel is integrated into the stack. The free expansion of the SOFC tubes is possible and helps to avoid a complicated design or a complicated and expensive matching of the used materials of all stack components. The short sealing length is a further benefit of any tubular design. The power density of the proposed tubular stack design can be expected to be as comparable with planar stacks. The use of a small number of components and only completely circular parts of the SOFC tubes helps to control the manufacturing cost. It seems to be useful to make further investigations.

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