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AN ASSESSMENT OF DEPOSITION FOR POWER-PLANT
MOLTEN-CARBONATE FUEL CELLS

Richard A. Wenglarz

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POWER PLANT MOLTEN CARBONATE FUEL CELLS

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ABSTRACT

Particulate deposition in molten carbonate fuel cell anodes is addressed for operation with future coal gasification power plants. Power plant systems factors affecting deposition are explored such as gas cleanup requirements for particulate removal and gasifier product gas composition differences for various gasifier types and operational modes (air blown versus oxygen blown). Effects of fuel cell characteristics (including average cell current density and fuel utilization) on anode deposition are also quantified. Particulate effects on molten carbonate fuel cell anode performance may not be as detrimental as perhaps perceived in the past. Gas cleanup to remove virtually all particles larger than one micron in diameter is expected to prevent or at least greatly reduce anode deposition. However, cathode deposition in molten carbonate fuel cells should be evaluated in the future since cathodes are likely more prone to deposition than anodes even though cathode channel particle concentrations are much lower.

INTRODUCTION

The maximum practical theoretical efficiency for heat engines is 40 to 50% due to Carnot cycle limitations resulting from initial conversion of fuel to heat. However, fuel cell electrochemical reactions directly convert fuel chemical energy to electrical energy and therefore are not Carnot cycle limited and enable much higher energy conversion efficiencies. While conventional coal-fired power plants typically provide efficiencies less than 40%, power plants utilizing molten carbonate fuel cells operating with gasified coal might provide overall efficiencies approaching 50% from coal to AC busbar power and consequently are being considered for the future. For these advanced power plants, product gases from the coal gasifier will be treated by cleanup systems to remove particulate and chemical contaminants prior to entering the fuel cells. However, cleanup systems are not capable of completely eliminating impurities so that the fuel cells will ingest limited quantities of particulates and chemical contaminants.

Although significant efforts have been conducted to evaluate molten carbonate fuel cell tolerances to sulfur,^(1,2) halogens,⁽³⁾ and trace chemical contaminants,⁽⁴⁾ there appears to be no experimental or analytical work reported in the open literature which evaluates tolerances to particulates. However, due to the potential for small particles with diameters less than five microns entering fuel cell anode pores with average diameters from five to ten microns, it has been suggested⁽⁵⁾ that the safe operating particle concentration for fuel cell applications should be essentially zero, but no experimental or analytical basis was given for this estimate.

The following reports an initial assessment of effects of particulates on molten carbonate fuel cells. These evaluations are conducted for several power plant configurations representing a range of promising gasifier processes and a range of fuel cell operating characteristics.

FUEL CELL POWER PLANT

System Configuration

Figure 1 gives a simplified schematic representative of a coal gasification/molten carbonate fuel cell power plant. For this plant, a coal gasifier produces a fuel gas which passes through a cleanup system to remove most of the entrained particulates and chemical contaminants. The cleaned gases enter the anode sections of the molten carbonate fuel cell bank. Figure 2 illustrates one cell of a fuel cell bank. Fuel gas flows through the anode flow channels and penetrates into the anode pores. Typically 80 to 90% of fuel reactants H_2 and CO in the gas react with carbonate ions in the electrolyte which occupies the pores to produce electrons according to the following reactions:

The oxidation of CO by Eq. (2) is in competition with the more favored water-gas shift reaction which converts CO to H_2 :

The $CO_3^{=}$ ions utilized in Eq. (1) and (2) have migrated to the anode through the electrolyte after being produced in the cathode. Products H_2O and CO_2 of the anode reactions exit the anode pores into the fuel channel gas stream.

After leaving the anode sections, the gas enters a catalytic burner where unreacted fuel is burned in air using a catalyst. The exhaust from the catalytic burner, which contains a significant amount of CO_2 , is combined with a recycled cathode gas stream and an oxidant gas stream, which contains a significant amount of O_2 . This combined gas stream enters the fuel cell cathode sections where the CO_2 and O_2 react to form carbonate ions using electrons which have been produced in the anodes and have traveled by way of an external circuit to the cathodes. The continuous flow of electrons from anodes to cathodes results in a DC current which is conditioned to produce AC power using inverters. Heat energy is extracted from the cathode section exhaust gases to produce electric power in the bottoming cycle using a heat

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Fig. 1 — Coal gasifier/molten carbonate fuel cell power plant schematic

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FIG. 2 - Molten carbonate-fuel cell section

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recovery steam generator and turbines driving electric generators. The major portion of gas from the bottoming cycle is recycled to the cathode section inlet and the remainder is exhausted to the atmosphere.

Coal Gasifiers

Several types of coal gasifier processes are candidates for operation with future molten carbonate fuel cell power plants. These include fluidized bed, gravitating bed, entrained flow, and molten salt gasifiers, each of which can produce different fuel gas compositions. Estimated representative gas compositions for these processes are shown in Table 1. These compositions are for gases entering the fuel cells at 10 atmospheres pressure and 650°C (1202°F) after humidification to eliminate (by the water-gas shift reaction) carbon particle formation.

Particulate Cleanup Systems

Cleanup of the coal gasifier product gases prior to entering the fuel cell banks will probably be accomplished either by cold gas cleaning using wet scrubbers or hot gas cleaning using ceramic filters for final stage particulate removal. Gas cleanup with cold gas scrubbers is commercially proven while hot gas cleaning with ceramic filters is in initial stages of testing⁽⁶⁾ and consequently has not been commercially demonstrated. However, if successfully commercialized, hot gas filtration would offer some significant advantages over cold gas wet scrubbing since thermodynamic losses due to cooling, cleaning, and reheating the gasifier product gases would not be incurred.

Figure 3 shows penetration fractions (mass of particles passing through divided by mass of particles entering) versus particle diameter for scrubber particulate removal systems. Two curves are given bounding the performance data measured⁽⁷⁾ for eight types of scrubbers operating at various commercial plants including two coal-fired boiler plants. Figure 3 indicates very low penetration fractions for particles with diameters greater than 1 to 3 microns so that virtually all particles above this size range can be removed from gases by scrubbers. However, scrubber performance decreases very rapidly below this size range so that

FIG. 3 - Range of particle penetration for scrubber gas cleanup devices

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TABLE 1

Representative Mole Fractions for Gases Entering Fuel Cells*

	Gasifier A Fluidized Bed <u>(oxygen blown)</u>	Gasifier B Gravitating Bed <u>(air blown)</u>	Gasifier C Entrained <u>(oxygen blown)</u>	Gasifier D Molten Salt <u>(air blown)</u>
H ₂	0.2062	0.1477	0.2048	0.1427
CO	0.0926	0.0664	0.0946	0.0675
CH ₄	0.0918	0.0488	0.0941	0.0434
CO ₂	0.2937	0.1456	0.2948	0.1578
NH ₃	0.0001	0.0004	0.0001	0.0003
H ₂ O	0.3124	0.1548	0.3050	0.1595
N ₂	0.0033	0.4363	0.0066	0.4287

* Data given in this table were derived using as source configurations the Westinghouse fluidized bed, METC gravitating bed, TEXACO entrained, and Rockwell molten carbonate gasification systems.

only a relatively small fraction of particles may be removed for diameters of a few tenths of a micron up to, depending on the scrubber device, about one to a few microns.

Experimental ceramic fiber hot gas filters have also demonstrated very low particle penetration fractions (less than 0.01) for particle diameters above about one micron.⁽⁶⁾ However, like scrubbers, their particulate removal performance starts to very rapidly decrease for smaller diameters. Since the particulate carryover from coal conversion processes ranges in diameter down to about 0.1 microns^(7,8) and scrubber and hot gas filters are expected to remove virtually all particles with diameters greater than about one to three microns, the following fuel cell evaluations are conducted for particle diameters in the 0.1 micron to 3 micron range. This particle size range of 0.1 to 3 microns where cleanup devices are least effective is also a range for which electrode pores (a typical average anode pore diameter is 5 microns) might be expected to be susceptible to particle blockage.

Fuel Cells

The conceptual molten carbonate fuel cell (illustrated in Figure 2) evaluated in this work has operating characteristics which are objectives of current development work⁽⁹⁾ for future power plants. The cell produces an average power density of 0.135 watts/cm³ (125 watts/ft²) of cell area at an 0.8 v/cell potential. For operation with future power plants, the anode inlet gas pressure and temperature are assumed to be 10 atmospheres and 650°C (1202°F), respectively.

As indicated by Figure 1, the gas leaving the fuel cell anodes are diluted prior to entering the fuel cell cathodes. Since this dilution results in substantially lower particle concentrations in the cathode flow channels compared to the anode flow channels, only anode deposition is addressed here.

PARTICULATE EFFECTS

Mechanisms of Deposition

The low flow velocities (typically less than 30 cm/sec (1 ft/sec)) in fuel cell channels is expected to cause negligible erosion. However, particles entrained in the gases flowing through electrode channels can potentially degrade fuel cell performance by i) depositing on channel surfaces to restrict flow, or ii) entering into and blocking pores. Pore blockage could potentially reduce cell output by inhibiting H_2 and CO fuel reactant migration into anode pores and H_2O and CO_2 reaction product migration out of the pores, driving reactions of Eq. (1) and (2) to the left (opposite to the desired direction).

Mechanisms of particle transport which affect delivery of particles to electrode surfaces and to the entrances of pores are:

- Brownian diffusion
- Electrostatic migration
- Gravitational settling
- Thermophoresis
- Convection

Due to the small cross section dimensions and low flow velocities for fuel cell electrode flow channels, flow is laminar and turbulent diffusion transport of particles is not in effect.

Particle delivery to anode surfaces and into pores have been evaluated for each of the transport mechanisms listed above. Brownian diffusion which transports particles by random impacts with thermally agitated gas molecules was analyzed using results⁽¹⁰⁾ for particle transport in laminar pipe flow. Electrostatic migration due to particles accumulating charges and being driven by the anode-cathode electrostatic force field was evaluated using relations⁽¹¹⁾ for levels of charges accumulated by particles. Gravitational settling rates were obtained by balancing gravitational and fluid friction forces on the particles. Thermophoretic delivery is associated with particles in a thermal gradient experiencing a net force in the direction from higher temperature to lower temperature due to higher intensity of bombardment by the thermally

agitated gas molecules on the hotter side compared to the cooler side. This effect was analyzed using results⁽¹²⁾ for thermophoretic forces on particles. Since fuel gas reactants H_2 and CO entering a pore have lower molecular weights than reaction products (produced according to Eq. (1) and (2)) exiting the pore, there results a net mass flow rate out from anode pores. Convection effects associated with this flow from anode pores influences delivery of particles to anode surfaces and their entry into pores. As will be described in more detail in the next section, flow rates from pores have been obtained by extending electrochemical models^(13,14) which have been developed to determine currents generated by individual pores.

Because the reactions of Eq. (1) and (2) release heat in anode pores which is predominantly absorbed by the metal (Ni) anode, the anode surface is at a higher temperature than the anode channel gas. As a result, thermophoresis, in addition to flow from anode pores acts in opposition to the other particle transport mechanisms which tend to deliver particles to the anode surface and pore entrances. Consequently, intrinsic characteristics of molten carbonate fuel cell anodes tend to counter deposition. The extent which deposition is alleviated will be addressed in the following sections. First will be described the results obtained for anode pore flow velocities which are necessary for these deposition assessments.

Pore Flow Velocities

As particles are carried parallel to the anode surface by anode channel flow and slowly descend toward that surface due to the transport mechanisms described above, those particles periodically pass through "jets" issuing from the anode pores. Each "jet" tends to slow, or even reverse, particle descent until it is traversed. Whether or not particles can reach the anode surface can be approximately addressed by comparing the near surface deposition velocities (relative to the flow) for the various particle transport mechanisms to the average flow velocity from the anode, assumed normal to the surface. If the average local flow velocity from the anode exceeds the local deposition velocity

then, based on first order effects, particles will not be able to reach the anode surface.

If the deposition velocity for a particle is sufficiently high that it can experience a net descent to the anode surface, it can deposit at locations between pore entrances or can potentially enter into those pores with low exit flow velocities. However, the particle will still be repelled from the entrances of those pores with sufficiently high exit gas velocities. Neglecting the fluid velocity distribution across the pore diameter, particle entry into a pore of significantly larger diameter than the particle will be addressed by comparing the near surface deposition velocities to the cross-section average flow velocity exiting the pore. If the average pore velocity exceeds the particle local deposition velocity relative to the flow then, based on first order effects, the particle will not enter the pore.

Determination of average flow velocities from individual pores and the local average anode flow velocity requires use of detailed information concerning anode characteristics which will only be summarized here. Electrochemical models for currents generated from individual pores and experiments on bench scale molten carbonate fuel cells have indicated⁽¹⁵⁾ that electrodes are permeated with the electrolyte by capillary absorption so that the smallest pores are filled with electrolyte (flooded pores) up to a diameter above which pores are virtually empty except for a thin electrolyte film on the pore walls (thin film pores). For efficient operation of molten carbonate fuel cells, only a small fraction (typically about 5%) of anode pore volume is filled and most of the anode current generated is by flooded pores.⁽¹⁴⁾ Consequently, a model^(13,14) which has successfully described electrochemical characteristics for flooded pores has been extended to determine flow velocities from pores considering the smallest pores are flooded up to a diameter (three microns for the cell analyzed) associated with 5% of total pore volume occupied by electrolyte.

Based on the above considerations, the average velocity U_p of flow from a pore of radius r has been found to depend on the local concentrations $C_{H_2}(x)$ and $C_{CO}(x)$ of fuel reactants H_2 and CO and local

gas density $\rho(x)$ at distance x along the anode flow channel from the inlet, average values $\bar{C}_{H_2}$ and $\bar{C}_{CO}$ of concentrations from the anode channel inlet to exit, average current density $\bar{I}$ over the total fuel cell area, and constants H and a involving anode characteristics and gas species molecular weights

$$U_p(x) = \frac{H}{r} \frac{\bar{I}}{\rho(x)} \frac{C_{H_2}(x) + a C_{CO}(x)}{\bar{C}_{H_2} + a \bar{C}_{CO}} \quad (3)$$

This equation shows that the average velocity of gas exiting flooded pores decreases with increasing pore diameter so that the potential for particle entry and blockage increases for larger anode pores. Flow rates from pores increase with increasing average current per cell area so that higher average cell current densities decrease the tendency for particles entering into and blocking pores. Because H_2 and CO concentrations vary for the output gases of the coal gasifier systems shown in Table 1, the potential for pore blockage is affected by the gasifier type and operating mode (e.g., air blown vs. oxygen blown). Since the concentrations of fuel reactants H_2 and CO decrease in progressing from the anode channel inlet to exit as the fuel is utilized, flow velocities from pores decrease along the channel length. Consequently, past regions of channel inlet flow disturbances which may enhance local deposition, the tendency for pore blockage continually increases from regions just past the channel inlet to the exit. The potential for pore blockage at downstream channel regions increases with increasing cell fuel utilization and corresponding decreasing exit concentrations C_{H_2} and C_{CO} .

The average local flow velocity from the anode has been obtained from dividing the mass flow rate from a local area by the product of that area times the local gas density. A similar relationship to Eq. (3) results except that the first H/r term is replaced by another coefficient H_a involving gas species molecular weights. Due to this similarity, effects on average anode flow velocity and the resulting inhibition of particle

arrival on the anode surface are related in the same way as described above to average cell current density, gasifier type and operating mode, location x from anode channel inlet to exit, and fuel utilization. Effects of these various molten carbonate fuel cell power plant system parameters on both particle delivery to anode surfaces and into anode pores are quantified in the following sections.

DELIVERY TO ANODE SURFACES

Figure 4 gives particle deposition velocities (mass arrival rates divided by gas stream particle concentrations) at the anode surface versus particle diameter for the various particle delivery mechanisms described above. The particle size range is that given in Figure 3 as likely to pass a power plant scrubber or filter gas cleanup system. Also indicated on Figure 4 are average flow velocities out from the anode surface at locations near the inlet and near the exit of anode flow channels for the gasifier coal gas compositions given in Table 1 and 85% fuel utilization in the anode channels. These values bound the decreasing average anode flow velocities over the channel length. As indicated earlier, based on first order effects, if this average flow velocity from the anode surface is greater than the particle deposition velocities to that surface, the net particle motion is away from the anode so that particle arrival is prevented.

For anode channel inlet regions, Figure 4 shows that the average anode flow velocity exceeds the particle deposition velocities for all delivery mechanisms and particle diameters of concern. Consequently, particle arrival to anode surfaces near the inlet is expected to be eliminated or at least substantially reduced. At regions near the anode channel exit and for particle diameters less than about one micron, both the average anode flow and thermophoretic effects which tend to drive particles away from the anode surface are about the same magnitude or greater than the other mechanisms which tend to deliver particles to that surface. Accordingly, delivery of particles with diameters less than about one micron to all anode surface locations is expected to be prevented or at least greatly reduced.

Curve 731162-A

Fig. 4 - Deposition velocities for molten carbonate fuel cells

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As indicated by Figure 4, the above results are not very sensitive to gasifier type provided that for each system the fuel cell is operated at the same average current density (in this case 0.169 amps/cm^2). However, utilization significantly affects the average anode flow velocities and the potential for particle arrival at downstream anode regions. Table 2 gives average anode flow velocities for the four gasifier types in Table 1 with the fuel cells operated at 80% and 90% combined H_2 and CO utilization. For all gasifiers, the average anode flow velocity near the channel exit at 90% utilization is a factor of about 2.7 lower than that for 80% utilization. The average anode flow near the channel inlet is not greatly affected by either gasifier type or utilization as shown in Table 2.

Figure 4 indicates that gravitational settling dominates all other particle transport mechanisms at locations near the channel exit for particle diameters larger than about one micron. Consequently, these larger particles can be delivered to the downstream anode surfaces. However, Figure 3 shows that the highest efficiency scrubber systems can eliminate from a gas stream virtually all particles with diameters larger than one micron. For less efficient scrubbers passing particles with diameters from one to three microns, the rates of particle delivery are so low as to restrict anode channel flow only over long durations. Even assuming a gas stream particle concentration of 5 ppm in the one to three micron range and that all arriving particles stay on the anode surface, the time to block 10% of an anode channel (of 0.25 cm (0.1 in) height) is projected to exceed three years.

DELIVERY INTO PORES

Prospects for particle arrival at anode surfaces have been discussed in the last section. The ability of arriving particles to enter into anode pores will be considered in this section. As described earlier, particle entry into pores can be addressed by determining whether the particle deposition velocity relative to the flow exceeds the flow velocity exiting a pore for pores significantly larger than the approaching particle. Figure 5 shows gravitational settling velocity versus particle diameter

TABLE 2

Average Anode Flow Velocities

<u>Gasifier</u>	<u>Utilization</u>	<u>Average Anode Flow Velocity at Inlet (cm/sec)</u>	<u>Average Anode Flow Velocity at Exit (cm/sec)</u>
A	80%	3.07×10^{-2}	2.17×10^{-3}
	90%	3.30×10^{-2}	8.07×10^{-4}
B	80%	2.92×10^{-2}	2.35×10^{-3}
	90%	3.12×10^{-2}	8.82×10^{-4}
C	80%	3.06×10^{-2}	2.15×10^{-3}
	90%	3.27×10^{-2}	8.00×10^{-4}
D	80%	2.88×10^{-2}	2.31×10^{-3}
	90%	3.08×10^{-2}	8.72×10^{-4}

compared to pore flow velocity versus pore diameter at regions near the anode channel exit where pore flow velocities are lowest. Deposition velocities associated with other mechanisms for particle delivery into pores are not shown because of their much lower values (by more than an order to magnitude) than pore flow velocities. Furthermore, as described earlier, for the particle diameter range below one micron where these other mechanisms are relatively important compared to gravitational settling, thermophoresis and the average anode flow are expected to prevent or at least greatly reduce the arrival of particles at the vicinity of pore entrances.

Figure 5 indicates that flow velocities from pores greatly exceed particle deposition velocities at all particle diameters less than one micron. Gravitational settling velocities approach and can even be greater than pore flow velocities for high fuel utilizations and particle diameters nearing three microns. Consequently, the blockage of flooded anode pores by particles of smaller diameters than the pores is in most cases prevented by the gas flow exiting the pores averting particle entry. Particle entry into flooded pores appears possible only for a worst case combination of particle diameters approaching three microns, high fuel utilizations and regions near the anode flow channel exit where pore velocities are lowest. Although the pore flow velocity curves in Figure 5 are for the gas composition of gasifier A, the above conclusions are applicable to all gasifier types in Table 1 since, as indicated earlier, results are not very sensitive to gasifier type.

The above results apply to flooded pores which have been shown to produce the greatest contribution to anode current. Because of larger diameters and apparently lower rates of fuel reactions, flow rates from thin film pores may be significantly lower than determined above for flooded pores. Accordingly, thin film anode pores may be prone to blockage by particles. However, the degree of fuel cell performance degradation due to whatever blockage that may occur is uncertain since the thin film pores are not the major contributors to anode current.

Fig. 5— Comparison of pore flow velocities with gravitational settling velocities

Although the above results indicate that particles smaller than one micron will not likely arrive on anode surfaces to block channel flow or approach pore entrances and that particles larger than one micron in diameter will not likely enter flooded pores of larger diameters, the possibility of particles larger than one micron alighting on and blocking the entrances of smaller pores has not been addressed. Whether or not the relatively high velocity but smaller "jets" issuing from the smaller flooded pores can reverse the relatively slow descent of larger particles requires a detailed theoretical analysis beyond the intent of this initial assessment of molten carbonate fuel cell deposition.

CONCLUSIONS

For future coal gasifier/molten carbonate fuel cell power plants, the effects of particulates on fuel cell anode performance may not be as detrimental as perhaps perceived in the past. Inherent characteristics of molten carbonate fuel cell anode operation tend to counter deposition. The most productive current generation areas of the anode are also the least prone to deposition.

Increasing average cell current density decreases the potential for fuel cell deposition. The potential for deposition is not very sensitive to variations in gas composition for a range of coal gasifier types or to air blown versus oxygen blown gasifier operation provided the cells are operated at the same average current density in each case. The possibility of channel flow restriction by deposition or pore blockage by particles increases for particle diameters greater than one micron, for increasing fuel utilization, and in progressing from the anode flow channel inlet to exit.

Due to gas flow from anode pores and thermophoretic effects, molten carbonate anodes appear capable of ingesting significant concentrations of particles with diameters in the range from one micron down to a few tenths of a micron which are most difficult to remove from gases using scrubbers, filters and other gas cleanup devices. Operation of future molten carbonate fuel cell power plants with highly efficient scrubbers or hot gas ceramic filters capable of removing from the coal gas virtually all particles with diameters larger than one micron is expected to result in very little, if any, blockage of anode flow channels or anode pores. (A possible exception may be at regions just inside anode channel inlets where flow entry effects could potentially cause local deposition.) For less efficient particulate removal allowing particles up to three microns in diameter to enter anode flow channels, slow rates of channel flow restriction due to deposition may occur at downstream anode locations. Particles with diameters greater than one micron but somewhat less than three microns may possibly alight on entrances of smaller pores but do not appear capable of entering flooded pores. However, particles in this size range may be

capable of entering larger thin film pores. Since these thin film pores are not the major contributors to anode current, the resulting extent of anode performance degradation is uncertain.

Molten carbonate fuel cell cathode deposition has not been considered in this work. Due to net flow of gases into cathode pores, molten carbonate fuel cell cathodes may be more susceptible to deposition than anodes, even though particle concentrations are much lower in cathode flow channels. However, it appears possible that cathode deposition could be reduced by judicious choices of cathode pore size distributions and cell operating characteristics.

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SUMMARY (Purpose, Scope, Approach, Results, Conclusions, Significance):

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