Development of ASME Design Basis for Reliable SOFC

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Motivation

- Provide recommended practices and associated modeling and analysis procedures for use by U.S. SOFC stack designers and fabricators
- Provide recommended practices for design of durable and reliable SOFC stacks
- Serve as a repository for state-of-the-art knowledge and experience gained in SOFC designs
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Approach

Create a document such that the body is organized in sections consistent with the workflow of the design process:

- High level material provided in main body
- Details of the scope and assumptions
- Technical details listed in Appendices

Design process workflow consists of:

- Development of the initial SOFC design envelope
- An iterative process that includes:
  - Analysis procedures and tools to create model geometries, setup, and perform simulations
  - Evaluate and redesign if necessary
    - Based on the Design/Failure criteria
  - Repeat
Work Flow

Scope and Assumptions → SOFC Requirements → Materials & Material Properties → SOFC Electrochemistry → Analysis Procedures and Tools

Analysis Procedures and Tools → Evaluation and Redesign

Evaluation and Redesign → Design Meet Criteria?

Design Meet Criteria? → Yes/No

No → Design Meet Criteria?

Yes → Design Meet Criteria?
Scope and Assumptions

➤ Scope
  - Provide a methodology for evaluation and optimization of SOFCs through modeling and analysis procedures

➤ Assumptions
  - Stationary system
  - Base loaded
  - Planar SOFC stacks
  - Steady-state operating conditions
  - Nominal component dimensions
SOFC Requirements

- Set of specifications generated by the overall system design that the stack must meet
  - Stack power output
  - Stack operating voltage
  - Fuel and oxidant flow specifications
  - Stack weight and volume
Materials and Material Properties

- Typical SOFC stack component material choices described
- Key material properties needed as input into the model or validation of model results provided, including the model it is applicable to:
  - Structural properties
  - Thermal properties
  - Electrical properties
  - Interfacial material properties
  - Fluid material properties
SOFC Electrochemistry

- **Elements of the electrochemical performance model (current-voltage relation)**
  - Nernst potential
  - The electrode reactions
  - Voltage loss terms

- **Current-voltage relation**
  - Describes the local performance
  - Applicable over expected range of fuel composition and utilization, temperature, and operating pressure
  - Calibrate with experimental data

- **Pressurization**
  - Performance boost with increased operating pressure
Analysis Procedures and Tools

► Physical phenomena being modeled
  ■ Mass transfer of fuel and oxidant fluids
  ■ Chemical and electrochemical reactions
  ■ Heat transfer
  ■ Solid mechanics

► Thermal-fluid-electrochemical analysis
  ■ Design considerations
  ■ Modeling procedure

► Thermal-structural analysis
  ■ Design considerations
  ■ Modeling procedure

► Modeling tools for SOFC
Design/Failure Criteria

Design criteria that must be met when identifying possible failure modes and associated mechanisms in SOFC materials and components

- Structural failure
  - Bulk material
  - Interfacial
  - Loss of contact
Evaluation and Redesign

► Review performance of the design and identify possible changes which have the effects:
  ■ Change(s) will help stack meet performance criteria
  ■ Numerical modeling can be used to ‘test’ the effect of change
  ■ Favorable changes have little or no impact on system or design process

► Redesign approaches ranked by impact:
  ■ Geometrical changes to components (least impact)
  ■ Stack operating conditions
  ■ Material modification/substitution (most impact)
Recent Progress

► Working meeting on May 30 - June 1 at PNNL
  ■ Focused on finalizing the document flow
  ■ PNNL, ORNL, and ASME participation
  ■ Obtained consensus on document modifications
  ■ Assigned authorship for various section rewrites

► Monthly teleconferences
  ■ PNNL, ORNL, and ASME participation
  ■ Using C&S Connect online repository

► Writing of document
  ■ Submitted second draft version of the document including sections 8 through 10 in June, 2008
  ■ Finalizing writing of Section 11 to complete the first document version
  ■ Presently under PNNL revision and internal review
SOFC Design Basis: Next Steps

- **Document writing**
  - Technical input for section components practically complete
  - Editing and reorganization of subsections and appendices
  - Ensure technical accuracy and completeness
  - Obtain NETL content approval
- **Final document assembly**
  - Ensure content and flow sufficient to convey design basis
  - Assemble ancillary information (material properties, examples, references)
- **Peer review**
- **Document “release”**
5.0 Scope and Assumptions

This document provides design guidelines for solid oxide fuel cells (SOFCs). These guidelines provide a methodology for evaluation and optimization of SOFC reliability, focusing on failure mechanisms associated with the thermomechanical properties and state of the SOFC.

SOFCs have been considered and designed for an extremely wide range of applications, from portable power units generating a few watts of power all the way to megawatt-scale central power plants. Each application comes with its own unique set of design requirements and challenges. This document addresses the design under steady-state operating conditions, but does not address long-term performance. These subjects will be addressed in another document. In order to simplify and focus this guide, the scope is limited by the following set of assumptions.

5.1 Stationary, Base-loaded Systems

The majority of current SOFC development is focused on large-scale (hundreds of kilowatts to hundreds of megawatts) central power generation. These stationary systems are planned to operate in relatively benign conditions. Specifically, they will be base-loaded, running near peak capacity at all times, with minimal turndown or load-following requirements. Shock and vibration are assumed to be insignificant contributors to the operating conditions for a stationary, base-load system. Start-up and shut-down rates can be as low as necessary, such that stresses during these operations do not exceed the stresses observed during steady-state operation or at room temperature when the system is shut down.

5.2 Design for Initial Operation

The design procedures in this guide have been developed to evaluate the performance of the SOFC during initial operation. The analysis conditions are representative of the operating conditions and the material state immediately after the SOFC system is placed in operation. As such, the evolution of SOFC components that may occur over long-term operation is not considered. Thermal transients associated with start-up and shut-down cycles and unplanned power outages (trips) are similarly beyond the scope of this document. An exception to this is that the room-temperature stress state associated with a fully shut down system condition are evaluated, as the SOFC will experience this state after manufacturing and prior to initial operation.

5.3 SOFC Stack Only

This document focuses on SOFC stacks, where the stack is defined as a single physical unit comprised of the fuel cell (electrolyte and electrodes), seals, fuel and oxidant flow-fields, interconnects and gas separators. In actual SOFC systems, stacks need to be integrated with other components, such as manifolds and current collectors. These components can be critical to SOFC performance and must be designed with care. Design guidelines for components outside the stack proper are beyond the scope of this document, however, and these components are assumed to minimally influence the behavior and stress state of the SOFC stack.
6.0 SOFC Requirements

As for all components, the SOFC stack is designed for use within a larger system. The system may contain a single stack or, more likely for central power applications, a large number of stacks arranged in a SOFC subsystem which in turn is a component of the overall power plant. In any case, the stack design must respond to a set of requirements or specifications generated by the system design. Typical SOFC stack requirements are listed here.

6.1 Stack power output

This is simply the gross electrical power generated by a single SOFC stack. In a system with only one stack, this value will be well-determined. In a large system with multiple stacks, trade-off analyses will typically be made between the overall system requirements and the stack manufacturing capability in order to establish the power of the individual stack.

6.2 Stack Operating Voltage

The operating voltage requirement will typically be driven by the needs of the system power electronics. A sufficiently high voltage will usually be required to achieve the highest possible dc-dc and/or dc-ac conversion efficiency. Note that since stacks may be connected electrically in series, higher overall voltages can be achieved without placing inordinate requirements on the number of cells in an individual stack.

The stack voltage and power output requirements, when combined with the expected performance characteristics of the fuel cells, will establish the size of the stack (i.e. the number and active area of the cells contained within it). The number of cells N in the stack is simply

\[ N = \frac{V_{\text{stack}}}{V_{\text{cell}}} \]

where \( V_{\text{cell}} \) is the nominal operating voltage for an individual cell in a stack of total voltage \( V_{\text{stack}} \). The active area of each cell is then related to the total stack power \( P_{\text{stack}} \) as follows:

\[ A_c = \frac{P_{\text{stack}}}{NI_{\text{cell}}} \]

where I is the stack current.

It is important to note that N and Ac will often be constrained by the capability of stack manufacturing processes and reliability considerations. As a result, the stack and system designers will not necessarily be able to exercise the freedom implied above in specifying stack power and voltage. At the present time, individual planar stacks are generally limited to \( N < 100 \) and cells to \( A_c < 1000 \text{ cm}^2 \). Again, series- and parallel-connected stacks can increase the effective values of these parameters.

6.3 Fuel and Oxidant Flows

SOFCs are driven by flows of gaseous fuel (typically a mixture of \( \text{H}_2, \text{CO}, \text{CH}_4 \) and various inert species) and oxidant (typically air). The system must provide specifications for these flows, while also leaving sufficient degrees of freedom for the stack design. The parameters to be specified for each gas stream are:

- Inlet and outlet temperatures
Table 7.1: Solid material properties required and the models to which they apply.

<table>
<thead>
<tr>
<th>Material Property</th>
<th>Model</th>
<th>SOFC Component</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Structural</td>
<td>Thermal</td>
</tr>
<tr>
<td>Young’s modulus</td>
<td>X</td>
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<tr>
<td>Poisson’s ratio</td>
<td>X</td>
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<tr>
<td>Coefficient of thermal expansion</td>
<td>X</td>
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<tr>
<td>Stress-strain curve</td>
<td>X</td>
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<tr>
<td>Thermal conductivity</td>
<td>X</td>
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<td>Emissivity</td>
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<tr>
<td>Interfacial strengths</td>
<td>X</td>
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<tr>
<td>Interfacial fracture toughness</td>
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<tr>
<td>Electrical conductivity</td>
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<td></td>
</tr>
<tr>
<td>Ionic conductivity</td>
<td>X</td>
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</tr>
</tbody>
</table>

7.1 SOFC Components and Common Materials Choices

7.1.1. Electrolyte

The electrolyte acts as a fast oxygen ion conductor, an electronic insulator, and gas transport barrier. To be effective, an electrolyte must be impermeable, possess sufficiently high mechanical strength to withstand fabrication- and operation-induced stresses, and possess thermal properties (e.g. thermal expansion) compatible with the other materials in the SOFC system.

In SOFC, yttria stabilized zirconia (YSZ) in the cubic phase is the electrolyte material of choice due to its fast oxygen ion conductivity, stability in reducing and oxidizing environments at temperature, and high strength. The most commonly used composition of this electrolyte is 8YSZ (8 mol% Y₂O₃ stabilized ZrO₂). Details on this and other electrolyte materials are given in Appendix A7-2.

7.1.2. Anode
8.0 SOFC Electrochemistry

The SOFC converts chemical energy into electrical energy by the electrochemical oxidation of hydrogen at the anode-electrolyte interface. Oxide ions from the cathode pass through the electrolyte to the anode/electrolyte interface where they combine with hydrogen gas from the anode to form water vapor and free electrons. The electrons freed by the reaction travel through the anode and an external conductor back to the cathode where the oxygen is reduced to oxide ions and the electrical circuit is completed. If carbon monoxide is present, it will also be oxidized and converted to energy. The maximum ideal open circuit potential is due to the difference in oxygen chemical potential between the cathode and anode sides of the electrolyte. This is the Nernst potential and can be expressed as a function of the partial pressure of oxygen in the anode and cathode:

$$V_{\text{Nernst}} = \frac{RT}{4F} \ln \left( \frac{P_{O_2,\text{cathode}}}{P_{O_2,\text{anode}}} \right)$$  \hspace{1cm} (8.1)

where $R$ is the universal gas constant (8.314 J/mol-K), $T$ is the temperature in Kelvin, and $F$ is Faraday’s constant (96485 A-s/mol). The Nernst potential for the combination of cathode air and wet hydrogen (ie. 97%H$_2$, 3%H$_2$O) on the anode is about 1.07 Volts at 750°C. The Nernst potential is decreased for fuels containing increased H$_2$O vapor pressure (such as reformed or recycled tail gas fuels), due to the subsequently increased O$_2$ vapor pressure in the fuel stream.

8.1 Fuel Cell Electrode Reactions

The SOFC can oxidize and consume carbon monoxide in parallel with hydrogen consumption. The following chemical reactions occur at the electrodes:

At the anode

$$H_2 + O^\cdot \rightarrow H_2O + 2e^-$$  \hspace{1cm} (8.2a)
$$CO + O^\cdot \rightarrow CO_2 + 2e^-$$  \hspace{1cm} (8.2b)

At the cathode

$$\frac{1}{2}O_2 + 2e^- \rightarrow O^\cdot$$  \hspace{1cm} (8.3)

Overall

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O$$  \hspace{1cm} (8.4a)
$$CO + \frac{1}{2}O_2 \rightarrow CO_2$$  \hspace{1cm} (8.4b)

When the external circuit is closed, an electrical current is generated based on the rate at which hydrogen and carbon monoxide are consumed according to equations 8.2a,b. With the generation of current, voltage losses are introduced, the sum of which, taken from the Nernst potential, results in the cell voltage. At low current density, electrochemical activation polarization losses are of primary importance. Voltage losses at intermediate
Data collected during pressurized operation of tubular SOFCs showed that the cell voltage increase was larger than that attributable to the increased Nernst alone. It was thought that the additional performance was a result of decreased activation and concentration polarizations \cite{od5}. In the case of an anode supported planar SOFC, the concentration polarization decrease for a 20-30 μm thick air electrode would be minimal. During operation with coal gas, which can contain large concentrations of water vapor, one might actually expect an increased anode gas concentration polarization under high pressure conditions. The activation polarization however, should be independent of electrode thickness. And it has been shown that the exchange current density, at the heart of cell activation, is dependent upon the oxygen partial pressure \cite{6z}. For the purposes of this discussion, the focus is placed on the activation polarization.

The Butler-Volmer equation is used to describe the activation polarization $\eta_{\text{act}}$ as it relates to the current ($j$) and the exchange current density ($j_0$).

$$j = j_0 \left[ \exp \left( \frac{2F\eta_{\text{act},e}}{RT} \right) - \exp \left( -\frac{\alpha_e F \eta_{\text{act},e}}{RT} \right) \right]$$  \hspace{1cm} (22)

where $F$ is Faraday’s constant of 96485 coulomb/mol (or A-s/mol). For the SOFC cathode, the charge transfer coefficient $\alpha_e$ is equal to 2. As such (22) can be written exactly as:

$$j = 2j_0 \sinh \left( \frac{\alpha_e F \eta_{\text{act},e}}{RT} \right)$$  \hspace{1cm} (23)

In a two-electrode activation polarization model, an expression for the anode is also needed. The anode charge transfer coefficient is equal to 1. When $\alpha_e$ is equal to 1, (22) can be approximated by (23). Thus assume both electrodes follow the form of (23). This is a reasonable assumption because, the error introduced by assuming the form is small, and the model will require fitting to data by use of adjustable parameters. Then the activation polarization for both electrodes is written as:

$$\eta_{\text{act},e} = \frac{RT}{\alpha_e F} \sinh^{-1} \left( \frac{j}{2j_0} \right)$$  \hspace{1cm} (24)

The exchange current density $j_0$ is dependent upon the oxygen partial pressure $PO_{2,e}$, expressed as \cite{7z}:

$$j_0 = \beta \exp \left( -\frac{E_{\text{act},e}}{RT} \right) PO_{2,e}^{-\frac{1}{2}}$$  \hspace{1cm} (25)
The electrochemistry model will require calibration to cell data to simulate SOFC stack performance. As an example, Figures 8.1a and 8.1b show the model compared to data in temperature and fuel concentration dependence respectively [3z]. The model simulates the cell performance for the full operating range of temperature and fuel concentration. This enables accurate calculation of the local current density, and source terms for the thermal-fluid solution, based on the conditions at each computational element of the planar cells within the stack.

**Figure 8.1.** Experimental and modeled a) temperature dependent, and b) fuel concentration dependent performance data for a given cell material.

### 8.3 Pressurization

When an SOFC is pressurized, its Nernst potential increases based on the partial pressure of oxygen at the cathode-electrolyte interface.
9.0 Analysis Procedures and Tools

9.1 Physical Phenomena Being Modeled

Numerical simulation of the SOFC requires the user to develop suitable models to capture the multiple interdependent physical phenomena which are occurring. Researchers have developed a large number of models describing vastly different aspects of SOFC operations from high level system models to very detailed atomistic studies. An engineering approach is taken in this guide, where simulations for the SOFC typically include four components: 1) mass transfer of the fuel and oxidant fluids, 2) chemical and electrochemical reactions to describe the conversion of fuels in the cell to electric power, 3) heat transfer to describe the thermal state of both the fluid and solid domains, and 4) solid mechanics to describe the mechanical deformations and stresses of the thermally-loaded structural components. In general, the fluid mass transfer, electrochemical performance, and resulting temperature field are so highly interdependent that they must be solved simultaneously within a single coupled model. However, these behaviors can usually be assumed to depend only on the initial stack geometry and be independent of any geometric deformations. Therefore, the modeling approach taken here is to compute the flow-thermal-electrochemical response and thermal-structural response independently using separate models which are sequentially coupled through the shared temperature field.

The fluid dynamics solution determines the pressure, velocity, and species concentration fields of the fuel and oxidant streams as they pass through the cell. For planar cells, the flow in the cell active area can be well approximated by a two-dimensional (2D) flow field with incompressible fluids and fully-developed laminar flow. The pressure drop can be calculated by assuming fully-developed laminar flow between parallel plates or through a porous media. Within a CFD-based framework, a solution for the full three-dimensional (3D) fluid domain can be obtained. Within a FEA-based framework, reduced order modeling is used for faster computations.

The electrochemical models have been described fully in Section 8.0. For planar cells in series, the local effects of interconnect/cell geometry on the electric field are ignored in this engineering approach. A cell is assumed to have the same potential over the entire active area, such that the electrical model for the cell consists of only a one-dimensional (1D) series of electronic and ionic resistors at each computational point on the active area. For several cells assembled in series, the entire stack is then a series of 1D resistors. Therefore, the current density is allowed to vary across the active area of each cell, but every cell in the stack has its own voltage and carries the same total stack current. The electrochemical model is also represented by a local 1D model. Species are consumed/formed at each calculation point on the active area according to the electrochemical reaction model, and the corresponding sinks/sources for the species balance are included in the fluid dynamics solution.

The thermal behavior of the stack involves the conventional heat transfer processes of conduction, convection, and radiation was well as reaction heat sources from the electrochemistry. The temperature field in the solid stack components is usually calculated for the full 3D domain to facilitate the subsequent stress calculations. The stack conducts heat through all of the joined components in the model according to their thermal conductivities. Convection heat transfer between solid and fluid domains occurs internal and external to the stack. Externally, the stack or its insulated enclosure will dissipate heat to the ambient and can
9.3.2.2. Fit model to data

The physics-based EC model has adjustable parameters to enable fitting of the model to data. The designer might begin with default (or typical) values for these parameters, then adjust as necessary to achieve sufficient fitting. In this approach, fitting the model to the data can be achieved by adjusting parameters $\alpha$ in the Butler-Volmer equation (Eq. 8.6) and $E_{\text{act}}$, the exchange current density relation (Eq. 8.7). Figures 10.1.1a and 10.1.1b show the effect that the parameters $\alpha$ and $E_o$ have on the model curvature. The combined effect of $\alpha$, and $E_o$, illustrated in Figure 10.1.2, gives an example of how the curvature of the performance curve can be manipulated to have the desired slope at a given current density. The pre-exponential ($\beta_{\text{act}}$) of eq. 8.7 is also adjustable to affect the magnitude of the exchange current density. Once the theoretically based electrochemistry model has been sufficiently adjusted to fit the experimental data, the model can be applied to simulate normal operating conditions of the stack.

![Figure 10.1.1. Performance curve manipulation by adjustment of the Butler-Volmer parameters a) $\alpha$, and b) $E_o$.](image)

![Figure 10.1.2. Performance curve manipulation by combined adjustment of the Butler-Volmer parameters $\alpha$, and $E_{\text{act}}$.](image)

In the event the performance data is limited – for example: to a single operating temperature and fuel composition – proceed with fitting of model to the available data.
9.4 Thermal-Structural Analysis

9.4.1 Design Considerations

9.4.2 Modeling Procedure

9.4.2.1 Material properties

The thermal-mechanical materials properties of each component must be defined for the analysis. The simplest constitutive model is linear elasticity with thermal effects, which minimally requires the elastic constants and a coefficient of thermal expansion (CTE). The elastic constants describe the stiffness of the material and consist of two of the following three quantities: elastic modulus $E$, shear modulus $G$, or Poisson’s ratio $\nu$, which are related by

$$E = 2(1 + \nu)G$$

Modulus data can be measured according to the procedures discussed in Section 7.0, or alternatively properties for many of the more common SOFC materials can obtained from the literature. The CTE describes the amount of thermal strain $\varepsilon^{th}$ in a component due to temperature change from the original temperature state $(T - T_{REF})$ often called the stress-free temperature. The CTE can be reported in the literature as an average ($\alpha$) or instantaneous ($\alpha_{inst}$) value according to the following relations

$$\varepsilon^{th} = \alpha(T)(T - T_{REF})$$

$$\varepsilon^{th} = \int_{T_{REF}}^{T} \alpha_{inst}(T)dT$$

The user must ensure that the CTE data is appropriately converted for the FEA code being used (see Appendix A9-4). Since the user is often interested in the operation stresses at high temperatures as well as the shutdown stresses at room temperature, the variation of the elastic constants and CTE must be accounted for whenever possible. Materials will typically have a lower stiffness at high temperature and CTEs often vary over the large temperature range being considered. The greater component compliance due to reduced material stiffness at elevated temperature will be important for the stress predictions. Elastic material properties are usually used for the first analysis of a new design. This is usually a good assumption for the ceramic components such as the cathode, electrolyte, anode cermet, and glass-ceramic seals. For ductile metallic components such as the interconnect or current collector, they may develop non-recoverable plastic deformations under high stresses. For these materials, an elastic-plastic material model with thermal effects is often used if the material data is available. The material model will consist of a yield stress to identify the onset of plasticity and a hardening modulus to describe the stress-strain response above the yield stress. The yield stress is based on a yield criterion (often the maximum distortion energy theory) that describes how the material tested under uniaxial loading yields under multiaxial stresses. A hardening rule is used to evaluate how the yield criterion changes after plastic strains are induced, but that is less important since the focus here is primarily on monotonic loading by the steady-state thermal field and stress reversal or cycling is unlikely. Once the material properties are obtained, they are assigned to
Figure A9-2.1. The main men in Mentat-FC.

Figure A9-2.2. Geometry setup menus and submenus.
10.0 Design/Failure Criteria

10.1 Bulk fracture

A discussion of structural failure of SOFC materials and components requires identification of the possible failure modes and mechanisms. In general structural failure in SOFCs can be categorized into cohesive and adhesive failure. Cohesive failure refers to the fracture of individual materials or components (e.g. electrodes, electrolyte). Adhesive failure refers to delamination that may occur at the interfaces formed by dissimilar materials or components in the cell or the stack (e.g. between the electrolyte and the electrodes, between the electrodes and the interconnects, between the seal and other cell components).

For each one of these failure modes several failure criteria have been developed. In the case of cohesive failure the four classical failure criteria include the maximum normal stress, the Tresca criterion, the Mohr-Coulomb criterion in which shear failure does not only depend on the shear stress but also on the normal stress and the von Mises criterion [Reference to Strength of Materials Book]. In the case of brittle materials, such as those used in SOFCs, where yielding and failure occur simultaneously due to lack of plasticity, the maximum principal stress criterion has been found to describe their failure adequately. The maximum normal stress criterion can be expressed as follows:

\[ \sigma_{\text{max}} \geq \sigma_o \]

where \( \sigma_{\text{max}} \) is the principal normal stress and \( \sigma_o \) is the strength of the material.

It is important to consider that the strength of brittle materials, such as those used in SOFCs, is stochastic and can be best described by a distribution (e.g. Weibull distribution). In this design guide the maximum principal stress failure criterion will be used to describe the failure of SOFC materials and a simplified approach will be used to account for the variability of strength data exhibited by SOFC materials.

An alternative approach to describe the failure of brittle materials is Griffith’s criterion, which is the basis of elastic fracture mechanics. According to this criterion a brittle material will fail when the stress intensity factor \( K_I \) reaches a critical value \( K_{IC} \). The relationships among the stress intensity factor \( K_I \), the far-field tensile stress normal to the crack, \( s \), and the crack half-size, \( a \), are given by

\[ K_{IC} = Y \sigma (\pi a)^{0.5} \]

where \( Y \) is a geometric parameter.