YSZ COATED FERRITIC STAINLESS STEEL

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ABSTRACT

Solid oxide fuel cells (SOFCs) can potentially extend the nation's energy reserves. However, to be reliable, the fuel cell chambers must be sealed to prevent leakage between the existing glass layers and the metal interconnects. InnoSense LLC (ISL) is producing a cost-effective thin yttria-stabilized zirconia (YSZ) coating system to provide a barrier layer between SOFC compliant glasses and, both bare ferritic stainless steel and spinel-coated ferritic stainless steel (Allegheny Ludlum SS441). This will result in high-quality YSZ coatings to SOFC interconnects for mass production by silk screening or spraying a metallo-organic solvent formulation, then drying and firing in commercially available thick film furnaces. Silk screening affords a low cost, fast design cycle technology that allows endless shapes to be successfully printed on a flat surface.

ISL is focusing on cost-effective application of thin, dense, adherent YSZ coatings on select portions (i.e., a square annulus) of SOFC interconnects.

ISL has developed a thixotropic formulation for screen printing and a non-thixotropic formulation for spray applications. In addition, the transparent glass-like films produced by firing these formulations will be rendered to fine micro-crystalline films, to prevent attack of the interconnects by the compliant glasses. ISL will conduct adhesion testing of our initial coatings on 430 stainless steel test plaques. YSZ coating applied to commercial Allegheny Ludlum 441 stainless steel will be subjected to compatibility testing by Fuel Cell Energy, Inc. (FCE).

BACKGROUND AND TECHNICAL APPROACH

There is a need to supply the Nation's electrical power needs using domestic energy sources. SOFCs are a part of reaching this goal. SOFCs offer clean, efficient, electrical power generation using multiple energy feedstocks. Their cost is reduced by using ferritic stainless steel such as Allegheny Ludlum 441 stainless steel (441SS) rather than costly ceramic conductive elements. Cracks, however, can occur in SOFCs at certain locations.

Recent developments of compliant glasses have addressed seal cracking, but these glasses interact with the stainless steel conductive elements. ***A thin, 1–2 µm, adherent Yttria Stabilized Zirconia (YSZ) coating can act as a diffusion barrier, but must be applied to shapes like a square annulus and be economical.***

ISL is accomplishing this by:

- **Using Organo-metallic and metallo-organics formulations for:**
  - Even coating of surface
  - Compositions that can be varied
  - Templates allowing complex deposition and shapes
  - Application by low cost industrial processes
  - Easily changed deposition shapes for low cost design changes
  - Overcoming classic difficulties with cracking over 1 µm thickness by formulation
- **Developing a screen printable ink.** Formulations were made from stable, metallo-organic/organo-metallic precursors. Initial formulations quickly precipitated to form YSZ powders. This difficulty was overcome by utilizing Hansen Solubility Parameters. Successful inks also contain appropriate ink additives. These inks can be printed using patterned stainless steel screens to deposit a square annulus on a flat 430SS or Allegheny Ludlum 441SS at low to high speed, for both low to high volume production,
- **Identifying screen and spray parameters** including: **Screens**: wire diameter, weave type, angle of weave, emulsion type, emulsion thickness, squeegee type and **Spray**: Paasche H spray machine for patterns up to 1-1/2 inches wide, spray head, pressure, number of passes, and distance from the substrate to achieve a good YSZ coating on 430SS and Allegheny Ludlum 441SS,
- **Investigating rolling or mechanical roll transfer** (High speed, high volume production, low material waste) – proof of principle single roll transfer,
- **Developing a firing profile** that will burn off organic material, nucleate YSZ crystal growth, and grow YSZ crystalline material from an initially transparent YSZ coating to produce a 1-2 µm YSZ layer,
- **Coating 4 in. x 4 in. test plaques of Allegheny Ludlum 441SS,**
- Having FCE perform **seal characterization** on 4 in. x 4 in. test plaques with & without spinel coating
- **Determining production cost** from raw material cost, capital equipment, floor space and burdened labor rate.

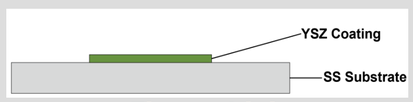


Figure 1(a)
Test plaque with YSZ coating prevents chemical reaction of Compliant Self-Sealing Glasses with metal conductive elements

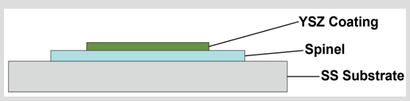


Figure 1(b)
Test plaque with YSZ coating on spinel coated 441SS prevents chemical reaction of Compliant Self-Sealing Glasses with metal conductive elements

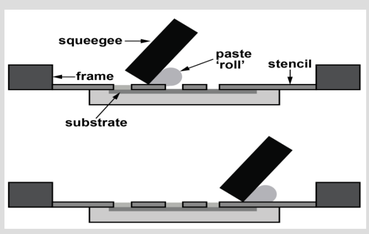


Figure 2
"On-contact" printing for low-medium volume production is more precise, and deforms the stencil screen less

EXPERIMENTAL SECTION

MILESTONE #1 – YSZ films deposited on 430 stainless steel plaques.

ISL has developed an initial YSZ metallo-organic based screen printable ink system. The ink is a modified sol-gel formulation. Since good printing inks require thixotropic rheology to induce leveling and prevent edge flow, a thixotropic agent has been included. A stress relief agent is also being incorporated into the formulation to relieve stresses in the deposition that would otherwise result in crack propagation in the films upon drying.

ISL is developing sprayable and screen printable formulations:

- **Photolithography used to make screen for printing**
- **Fired in air or controlled atmosphere**
- **Fired on stainless steel**

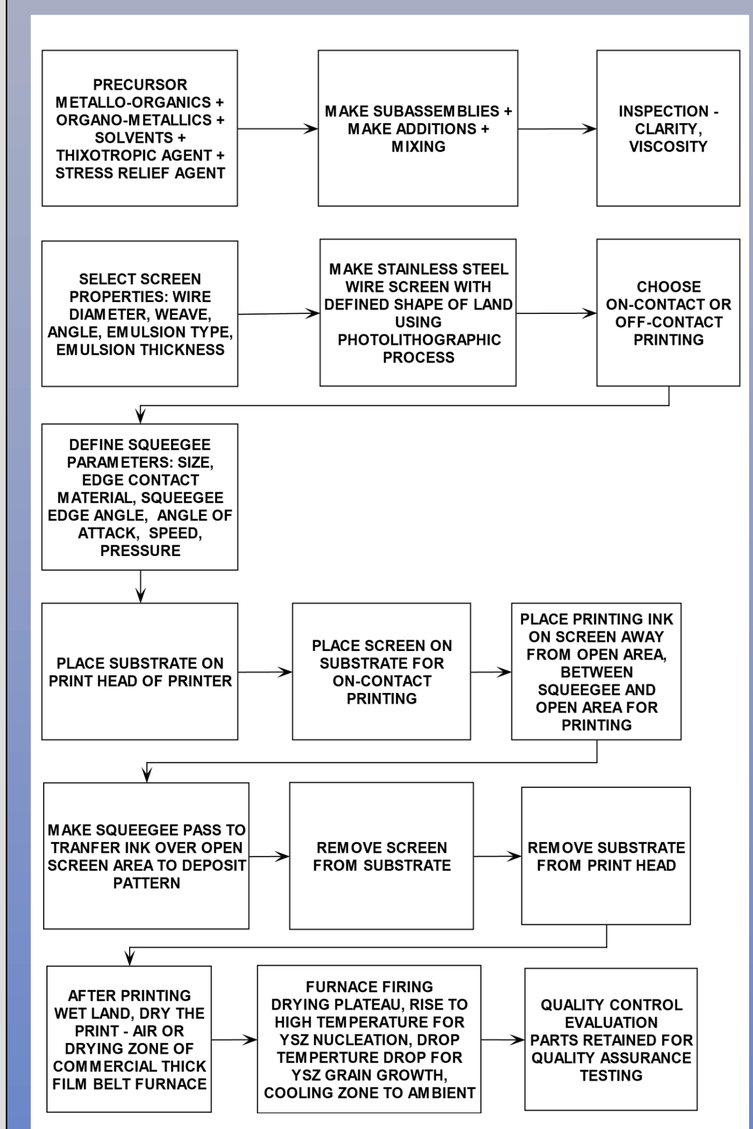
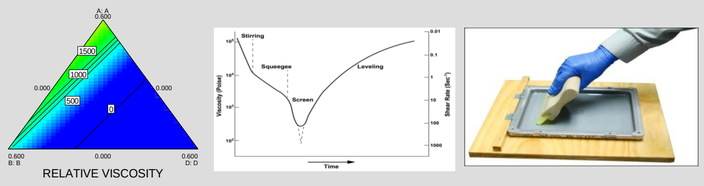


Figure 3.
Flow chart for screen printing YSZ patterned stencil coating substrates 441SS

RESULTS

YSZ Barrier Layer prevents chemical reaction of Compliant Self-Sealing Glasses and metal conductive elements.



(a) **(b)** **(c)**

Figure 4
(a) Optimization of variables is carried out using Statistically Designed Experiments, resulting in predicting response variables, such as, relative viscosity, **(b)** targeting typical screen printing viscosities[1] and **(c)** manual printing with stainless steel screen with print pattern.

1. Holmes, P. J. and Loasby, R.G.; Handbook of Thick Film Technology; Electrochemical Publications, Ltd.; p.5, 1976.

RESULTS (cont'd)

MILESTONE #2 – Characterization of YSZ films on 430 stainless steel plaques.

Conclusions:

- **Low cost, high volume standard film deposition techniques**
  - **screen printing**
  - **spraying**

(a) **(b)**

Figure 5
Comparison between **(a)** screen printed and **(b)** sprayed 0.5 in. x 0.5 in. pattern on 430 Stainless Steel

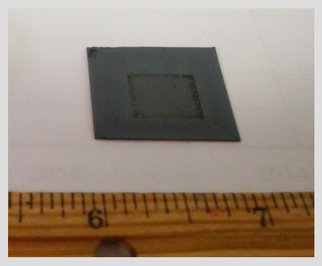


Figure 6
Sprayed 0.5 in. x 0.5 in. YSZ coated 430SS 800 °C firing in N_2- cooling from 650 °C to ambient temperature in air.

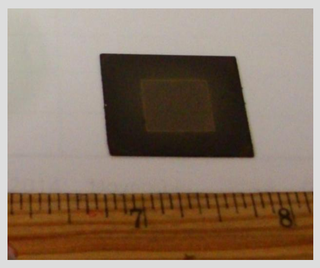


Figure 7
Screen printed 0.5 in. x 0.5 in YSZ coated 430SS fired 800 °C firing in N_2 to ambient temperature.

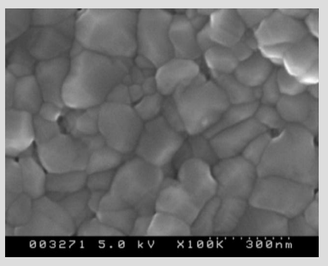


Figure 8
Screened 0.5 in. x 0.5 in. YSZ coated 430SS 800 °C firing in N_2- cooling from 650 °C to ambient temperature in air.

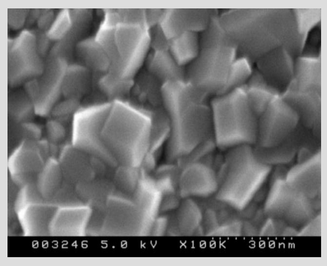


Figure 9
Screened 0.5 in. x 0.5 in. YSZ coated 430SS 800 °C firing in N_2 to ambient temperature.

PROJECT SCHEDULE AND MILESTONES

The following milestones will be accomplished in **Phase I**: **Phase I Project Tasks, Timeline and Milestones**. The major tasks and milestones for **Phase I** will be accomplished in accordance with the schedule shown in Figure 10.

Major Tasks	Months After Project Initiation								
	1	2	3	4	5	6	7	8	9
1. Finalize Performance Goals									
2. Acquire Material and Equipment									
3. Formulate SOFSeal™ Coatings and Apply to Stainless Steel Plaques									
Milestone 1: YSZ films deposited on 430 stainless steel plaques									
4. Characterize YSZ Coated 430 Stainless Steel Plaques									
Milestone 2: Characterization of YSZ films on 430 stainless steel plaques completed									
5. Test YSZ Films on Allegheny Ludlum 441									
Milestone 3: YSZ deposited on Allegheny Ludlum 441 stainless steel evaluated by FCE									
6. Production Cost Assessment									
Milestone 4: Production cost assessment completed									
7. Evaluate the Product's Commercial Potential									
8. Submit Reports									

Figure 10
Research Program Schedule for Phase I.

Work Accomplished to date: Formulations have been developed for both screen printing and spray application. Initial problems with precipitation of YSZ were eliminated, with inks stable at least for 2-3 months. A thixotropic additive was successfully incorporated into the formulation, along with a stress relief agent to eliminate drying cracks. Prints show good edge definition, without screen mesh marks. Printed and sprayed 1 in. x 1 in. samples on 430SS were fired up to 800 °C and show good adhesion. Also, the formation of crystalline material is in the range of 0.03 – 0.15 µm diameter. Additional improvements and testing are in progress.

Acknowledgments

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