Rapid Prototyping of PEM Fuel Cell Bi-Polar Plates Using 3D Printing and Thermal Spray Deposition

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Abstract

This article presents the results of exploratory research on novel methods for the fabrication of functional, metallic, gas flow, bi polar plates (BPP) for use in proton exchange membrane fuel cells (PEMFC). Low cost, high speed, additive manufacturing methods that combine 3D printing (3DP) and thermal spray (TS) technologies are described. Functional flow plates were manufactured by creating 3DP patterns and then depositing, and releasing, dense metals with TS methods. The new method yields dense metal plates, with interesting options for material choices and complex designs.

1.0 Introduction and Prior Art

The development of hydrogen fuel cell technology is highly relevant to current U.S. industry, global politics and environmental preservation. Gas flow plates are at the core of proton exchange membrane fuel cell (PEMFC) operation, and they present significant manufacturing and design challenges. The gas flow plate is tasked with delivering reaction gasses to adjacent membrane assemblies of a fuel cell, conducting the generated electricity, removing the H_2O byproduct, and influencing stack temperature.

There are three main established methods for manufacturing flow plates: (1) subtractive machining of graphite plates, (2) molding graphite or carbon materials, and (3) stamping metals. These three current industrial methods are limited by slow, expensive subtractive manufacturing methods, either directly at the part level or indirectly at the tool creation stage. Investigating high-speed, low-cost, additive PEMFC manufacturing methods may provide a superior path to the deployment of renewable, zero noxious emission energy sources.

1.1 Machined Graphite Plates

The first widely used method for creating research-grade gas flow plates, machining them from graphite plates, is only suited for low-volume production. All of the plate’s functional geometry is created via CNC machining. An example of a machined graphite plate can be seen in Figure 4.

In order to avoid even higher machining costs, flow plate design has been limited to comparatively simple planar geometry, with uniform features typically on the order of 0.79mm in width and depth. These feature dimensions appear to be dictated by existing machine tool design and are not inherently optimal for PEMFC performance. This per-part machining is very expensive and does not scale well with the eventual, requisite high-production volumes associated with the transportation industry.

1.2 Molded Graphite and Carbon Plates

Less expensive techniques for the manufacturing of graphite plates were researched by numerous investigators [10]. Thick graphite mats and chopped carbon slurry materials are made for companies such as Ballard Power Systems Inc. by Graftech and others. The mats or slurry are molded inside a die under elevated pressure and temperature. These moldable graphite materials offer identical or only slightly degraded performance from their costly predecessors.

Similar to their machined predecessors, molded flow plate designs have been limited to comparatively simple geometry due to the cost and complexity of molding tool creation. Molded plates suffer from high minimum thickness requirements, which increases the stack volume and weight and, in turn, decreases stack efficiency.

1.3 Stamped Metal Plates

Flow plates made of metal are ideal in terms of electrical conductivity, porosity, thermal conductivity, formability, strength, weight and cost,. These obvious advantages have been apparent for several years and research has been done in this direction [1] [9] [19]. Another area that provides potential savings is in the recycling of fuel cells after their product life cycle has ended [3], and in this respect metallic plates compare favorably to other materials. The primary drawbacks in using metals are corrosion resistance, tooling costs, and subtractive limitations.

Research has shown that plates made from 316 stainless with minimal finishing can perform nearly as well as machined graphite plates for a short period of time [3]. The decrease in performance is due to byproducts of metal corrosion inhibiting water transfer within the PEMFC membrane [12]. A coating, or some other corrosion resistance strategy, is needed for any affordable, non-noble metal flow plate.

Literature and patent reviews indicate that the corrosion problem with metal gas flow plates has received a significant amount of attention [1] [15] [16]. While specific solutions to this problem are closely guarded, the viability of these methods has been publicly demonstrated by the General Motors and Honda Motor Company [14] fuel cell programs, both of which use metal gas flow plates. A novel strategy for corrosion resistance for metal plates has been investigated within this exploratory project.

2.0 Motivation and Process Advantages

This research aims to address the aforementioned issues by investigating low-cost, additive prototyping and manufacturing methods. Reducing PEMFC cost, increasing stack design freedom, and accelerating the design cycles in fuel cell experimentation are the motivating factors behind this exploratory research.

2.1 Reducing PEMFC Costs

The current cost of PEMFC is still far above the $35/kW target established by the Department of Energy Future Car Congress, in fact, the current cost for PEMFC production is approximately $120/kW [4]. To be competitive with internal combustion engine performance a stack generating 80kW or more is needed, which means that even if the $35/kW target is attained, a stack would cost $2800. To put this figure into context, after 100 years of constant improvement, modern internal combustion engines sized for the same duty cost just under $1,000 to produce. After the membrane assembly, the most costly component of a PEMFC is the array of flow plates. Therefore, manufacturing optimization in this area can yield immediately useful results.

For the reasons cited above, we have chosen to investigate the production of metal flow plates. Production tooling for stamping, casting or molding made by traditional means offers a very cost-efficient method for producing metal plates, but it is expensive and slow to develop such tooling [2]. Not only is cost for tooling high, but the long term cost incurred through rework of part designs and tooling is also expensive. One way to reduce the costs incurred through the rework of part designs is to increase the amount of functional testing with prototypes, which our method allows us to create inexpensively. .

2.2 Existing Low-Price Technology and Materials

The two additive systems and the materials used in this research can be and sourced from existing companies and are also low in cost when compared to equivalent systems. The sample plates for this exploratory process were made for under $5 each, one at a time. Another cost advantage of an automated TS deposition process for manufacturing, when compared to other metal forming operations, is that there is comparatively low material waste. By depositing the part directly, the amount of process-induced waste can be significantly reduced, along with the total number of steps required for manufacturing.

2.3 Similarity between prototype and production

Although one target production method is the creation of production tooling for stamping, casting or molding of flow plates, there is the possibility that the methods we have developed to create prototypes can be scaled up to create production parts. Besides allowing for the differences in pattern material used, the same part materials can potentially be used for both prototype and production parts, by the same equipment.

2.4 Increased Design Freedom and Testing

Besides reduced costs and facilitating faster fabrication times, the geometric freedom provided by modern additive fabrication techniques can facilitate more advanced flow plate designs than can be achieved with the current subtractive methods. The existence of advanced stack designs in the literature but conspicuous lack of physical examples [11] illustrates the limits present in current PEMFC manufacturing. Solid oxide fuel cells (SOFC) have already incorporated non-planar designs [12]. With more flexible cost-effective manufacturing, PEMFC may also explore non-planar geometries.

Morgan Fuel Cells Inc. has done research into planar flow paths generated by computational fluid dynamics (CFD) simulations resulting in a claimed 16% increase in stack efficiency [17]. Such improvements could be built upon by investigating the incorporation of intra-channel geometrical features and non-planar cell designs. Also, by creating a faster method of functional prototype creation, different plate designs could be incorporated into the same PEMFC stack, to engineer an even pressure drop across the stack. This research into different plate combinations will require a cost-effective means to create complex functional parts for testing.

The ability to design, create and test a new, metallic gas flow plates design in a single day's time would be valuable those involved in PEMFC development and research. In addition to the speed of this novel process, the geometric complexity and freedom allowed for by the use of additive manufacturing will become of greater value and necessity as PEMFC stack design increases in complexity.

3.0 Materials and Methods

After a literature review and general problem investigation were completed the 3DP and TS process previously established by the researchers [21] were examined for BPP application. Figure 1 shows generally established targets for BPP properties [13]. Three serial preparatory goals were then set for the PEMFC investigation: prototype and permanent tooling design and fabrication, alloys selections, thermal spray equipment identification. Plate fabrication tests were then conducted after all preparations had been made.

BPP Material Property	Reason and Target
Permeability for gas	Separate anode and cathode
Electronic conductivity	$\geq$4 A cm^{-2}, <1 mV loss per plate
Density	Stack weight, <1 kg/kW
Thermal conductivity	Remove heat, 1 W per cm2 of MEA
Corrosion resistance	Proton activity of 1 M H_2SO_4
Pattern definition	High, equal pressure drop in all cells
Thermal & mechanical stability	Gas tightness, 5000hr. @ 80°C
Fabrication / Machining	DOE system target $45/kW total

Fig.1. Generally accepted performance targets for PEMFC BPP

3.1 Previously Established TS-3DP Process and PEMFC Specific Accuracy Test

A method for bonding and net-shaping TS deposited metals to 3DP patterns was previously investigated and described [21]. A series of alloys were deposited with TS and bonded to standard, commercial 3DP gypsum material. With high temperature stability and low cost, this existing material has been found suitable thus far. Up to 6mm of metal can be deposited in this type of "net-shaping" operation. Net-shaping in the thermal spray industry is typically done on engineered metal or glass substrates that are removed either through mechanical, chemical or other means. Gaining the ability to bond thermal sprayed coating to the target substrate was the first step towards the eventual PEMFC application. Figure 2 shows a variety of metal coated 3DP parts created at UofM.

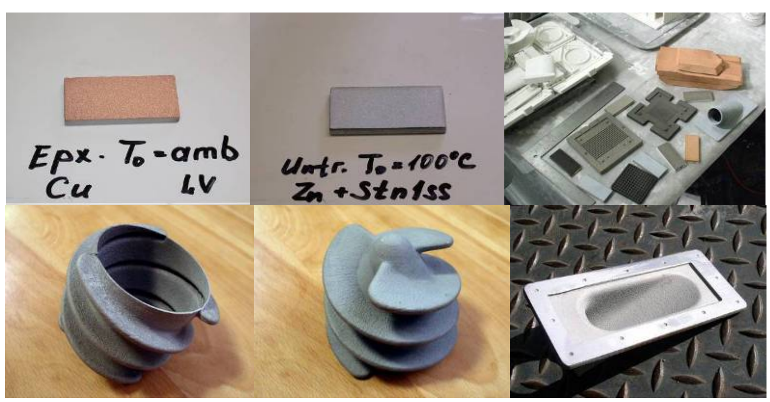

Fig.2. Initial TS-3DP compatibility test tokens, and an example of a net-shaped metal part.

With typical dimensional accuracy at +/- 250µm [22] current, commercially available 3DP machines were estimated to be capable of creating BPP geometry. While at the upper bound of acceptable accuracy tolerance, the low cost of the plaster based materials and high speed of the systems made them attractive for the proposed work. Feature accuracy tests were performed by the researchers on the 3DP machine used (ZCorp Z310). A resolution test token, with square geometry features ranging from 1mm to 100μm was created and a TS coating of Zn applied. 500μm channels were observed with optical microscopy, as shown in Figure 3. New 3DP machines with better accuracy and resolution are already available therefore, the tolerable accuracy of the 3DP method, was not seen as a problem in the short or long term.

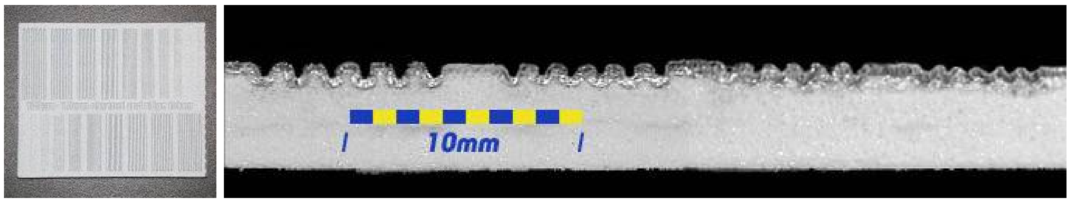

Fig.3. Resolution test token under optical microscopy shows ≥500μm square channels.

3.2 Prototype and Permanent Patterns Created

After basic 3DP and TS parameters were established, a single machined graphite BPP was purchased for reference, as shown in Figure 4. The sample plate is of very typical, basic design and was reverse engineered to serve as starting point for TS-3DP plate creation. The use of CMM and 3DCAD allowed for quick design work of research parts and tools. After the BPP CAD file was created, inverted-feature patterns were designed as tools. The first pattern was designed for manufacturing as a "green" 3DP part and the second pattern was designed as a 3DP investment cast H13-steel tool.


Fig.4. From left to right, benchmark graphite plate, CAD of 3DP pattern, sacrificial 3DP pattern, CAD of H13 permanent pattern, RP-IC cast H13 tool steel pattern..

The second pattern designed was created via an RP-investment casting process in H13 tool steel. This low cost, fast turnaround time method of geometrically complex, permanent tool creation was first employed by the researchers for use in casting applications [21]. The creation of the metal tool was done to demonstrate how this method of BPP fabrication can be employed in a larger production scale and not only for prototyping, as the 3DP casting patterns for the metal tools were made on the same type of machine that created the prototype patterns. An integrated cooling system and other complex features can be designed into a tool produced with these methods at no penalty in cost or production time.

The H13 patterns, as seen in Fig. 4, were cast by Weir Specialty Services of Houston Texas for a cost of under $1,000. These permanent tools were created in a few weeks time, concurrent with 3DP pattern usage. A BPP created from the H13 pattern requires the use of a release coating and compensation for thermal stresses for deposited metal part removal. Effort and decisions were made to keep the number of processing steps as few and as simple as possible, to better lend itself for future implementation.

3.3 Alloy Selection

The selection of alloys to be deposited for the initial exploratory tests was based on several factors and literature review [1] [7] [8]. Cost, TS compatibility, toxicity, melting temperature, reactions with atmosphere during spraying and PEMFC specific corrosion resistance, and compatibility with commercial coatings were the primary considerations. The ability for thermal spray equipment to deposit rapidly solidified, high temperature materials, such as cobalt and molybdenum was also considered. While such materials are not viable to consider in forming, machining or casting operations, aerospace and other industries have made good use of them as applied by thermal spray methods. This, and the ability to spray other, non-metallic materials onto near arbitrary geometry are key advantages that thermal spray brings to this application. For the initial trials, a 90-10, nickel-aluminum alloy was selected for the plate substrate and molybdenum was applied as a corrosion resistant layer.

Molybdenum's 2617˚C melting temperature is overcome by the TS equipment's ability to melt and deposit small amounts of material as it is deposited on the part. The corrosion resistant nature of Mo, particularly to hydrogen rich environments, makes it suitable as a coating for less expensive backing materials such as Ni. While the cost of molybdenum is higher than other common metals it is still reasonable to consider for automotive use. Considered in terms of a market price of \$70/kg for 99.7% pure Mo and the DOE established \$45/kW price target, a 0.2mm Mo coating for a fuel cell with a $1W/cm^2$ density would cost \$1.43/kW [5] [6].

3.4 Thermal Spray Equipment Selection

Concurrent with the development of the pattern creation strategy, the choice of thermal spray process was made. Conventionally, high performance coatings have been deposited either by plasma-based (APS) thermal spraying process, or by high velocity oxy fuel (HVOF) thermal spraying. Nevertheless, as compared with arc-spray process, the APS and HVOF processes usually have much higher cost and application difficulties. A recent development at the UM RML [23] [24] called the "HVOF/arc hybrid" gun, shown in Figure 5, was used to deposit the coatings. The hybrid process combines electric arc and HVOF spraying; molten metal produced by the arcing of wires is atomized and rapidly propelled to the substrate by a HVOF jet. This so-called "hybrid" concept offers many advantages over conventional equipment.

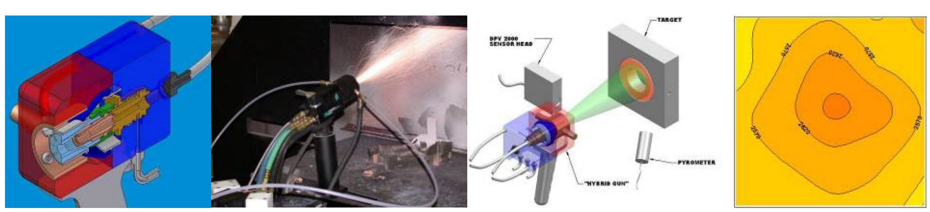

Fig.5. From left to right: hybrid arc-HVOF gun schematic , arc-HVOF photograph, sensor schematic and plume temperature characterization data.

3.4 Creation of Metallic bi polar plates

3DP patterns were fabricated and the metals for the BPP were deposited with the hybrid arc-HVOF equipment, creating metallic gas flow plates. The design of a 3DP pattern for use with TS metal deposition must accommodate thermal and dimensional factors that are introduced by the rapid solidification of the sprayed metal.

While not difficult to accommodate for once known, the ascertainment of thermal spray plume parameters required the use DVP/CMS 2000 measurement sensor made by TECNAR, Canada, as shown in Figure 5. The particle velocity and temperatures were measured at the target distance system enables the on-line measurement of surface temperature, velocity and diameter of individual particles in a spray plume by the principle of two-wavelength pyrometry [25].

Lower melting temperature metals such as Al and Zn can be sprayed onto 3DP substrates with relative ease. If spraying higher temperature materials, such as Mo with a melting temperature of 2617˚C, care must be taken. While the patterns remain intact when subjected to “over-temperature” spray, the pattern and deposited part can distort due to residual thermal stresses. The rapid solidification of metals during the thermal spray process can, however, offer unique advantages if controlled.

The 3DP patterns are sacrificed immediately after the plate’s metal has been deposited in the defined shape. The 3DP pattern is removed by an agitated soak in warm, soapy water, example 3DP-TS plates are seen in Figure 6. The time for pattern removal is relative to substrate dimension, but typically the process is complete in ten minutes. Four BPP patterns made with these materials required approximately 20 minutes to fabricate at a material cost of under $5.00 each. This low cost and quick fabrication time makes the process ideal for functional testing of fuel cell components. After the process parameters were established, the process was repeated to create multiple plates.

The trial plates made for this exploratory process were of bi-metallic with a Mo-Ni combination Manual spray of the sample plates required approximately 30 minutes, to create plates 1-2mm in thickness. Specific features of the plates created are described below.

4.0 Results and Discussion

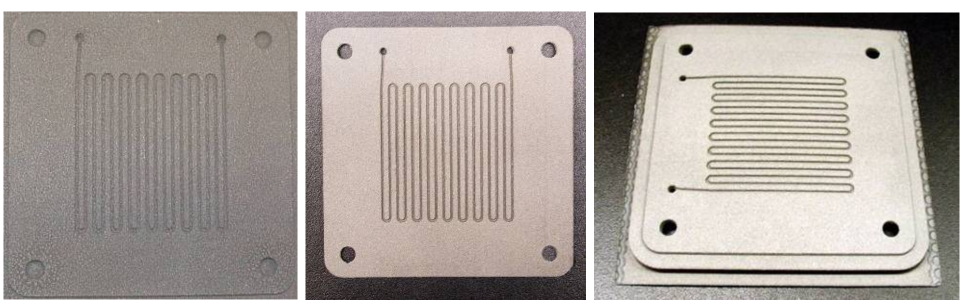

Fig.6. Sample plates created with the TS-3DP method

4.1 Dimensional validation

The completed, prototype gas flow plate, made on a 3DP pattern was measured for geometric accuracy. The part was digitized with a Perceptron Inc. laser, non-contact, 3D digitizer. The sprayed part’s outer dimensions were consistent with the accuracy of the 3DP machine used, at ≤250μm, as tested previously and as is seen in literature [22].

Internal channel features were consistently accurate to +/- 110μm in the X and Y axis, the Z axis exhibited material buildup that was due to manually spraying of the test pieces . There was a visible warp observed in the center of the plate in the Z axis, seen in blue in the CAD-to-part comparison in Figure 6. This was certainly caused by thermal conditions associated to the manual spraying of the part. As noted earlier, automation is used in the TS industry to facilitate high part accuracy and consistency.

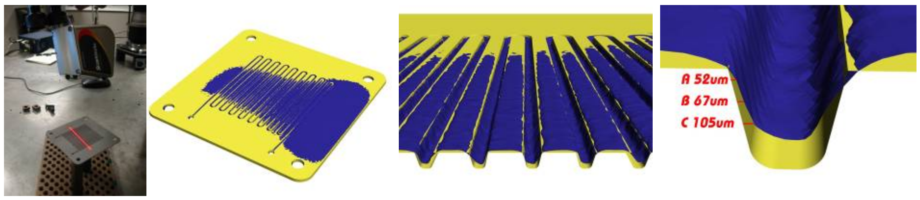

Fig.7. 3D laser scan, two CAD to part comparisons and in channel measurements

4.2 Metallography

The deposited metal from 3DP and metal tools was inspected in cross section with optical, atomic force and scanning electron microscopes to determine structure at several different scales Typical microstructure of the sprayed molybdenum is shown in Fig.   It is evident that the coatings were fairly dense without interconnecting porosities. Some very fine (~1-2 micron) dispersed pores were present in the microstructure, however, they don't pose any harm in terms of performance. Since the molybdenum layer is designed to prevent corrosion and gas penetration, it is important that the layer be dense.

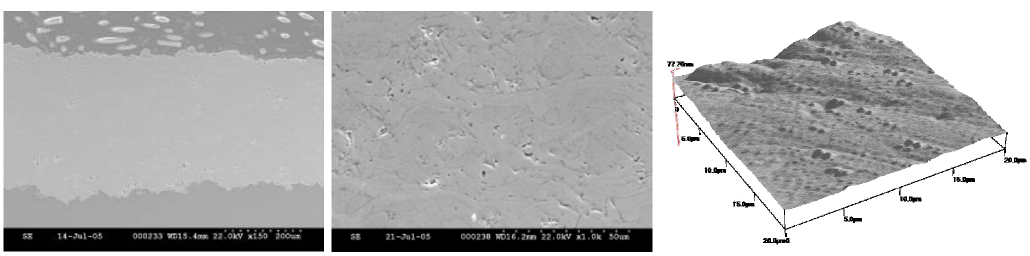

Fig.8. Cross section optical micrograph of TS deposited Mo on a wrought aluminum, sandblasted substrate with SEM and AFM

4.3 Mechanical Properties

The mechanical loads placed on BPP in a modern fuel cell are dominated by the varying compressive forces applied through whole stack compression, at different stack temperatures. While net shaped TS deposited metals in some cases do not retain all of the properties of their wrought equivalents, they do maintain high compressive strength and the choice of Mo as part of the bi-metallic construction offers very high strength to begin with.

4.5 Conductivity analyzed with STM

Conductivities on the sample were measured by a scanning tunneling microscope (STM). As well known, the current in STM depends on the tip separation as well as the inherent conductivity of the material. The tip separation depends on the sample surface roughness. Therefore, samples were polished by 1200 grit SiC paper prior to measurements. Measurements were taken over an area and averaged to get a comparative value since the surface asperity factor cannot be eliminated. Measurements were taken on copper, aluminum, sprayed molybdenum/aluminum and molybdenum wire. Figure below shows that coated molybdenum had a higher conductivity compared to molybdenum wire. Understandably, cold worked Mo has low conductivity due to increased level of lattice defects.

This method of point specific conductivity testing is superior to standard, bulk conductivity testing when doing FC analysis [20]. This is due to the fact that within a fuel cell current is being generated across the cell's active area, in very close proximity to the BPP. With this method, point specific conductivity can be observed and quantified relative to local plate features.

Figure 9 shows the average currents through various materials, this figure represents the average current of measurements taken from a range of probe distances. Therefore the overall conductivity of the area tested can be seen as the integral of the curve in Figure 9. It is to be noted that the bimetallic Mo/Al coating had reasonably good current compared to aluminum only. This demonstrates that thermal sprayed plates would not suffer from lack of conductivity.

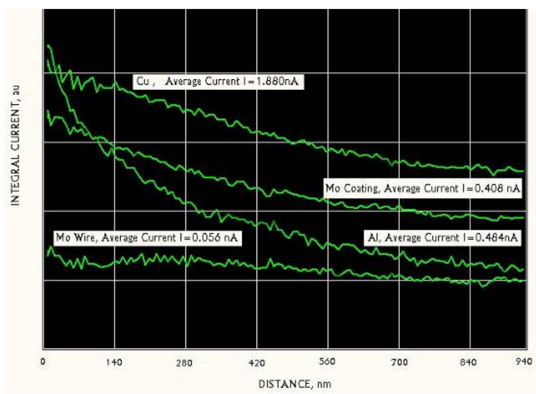

Fig.9. Current at probe-distance-from-sample is shown for multiple materials, as it can be seen the Mo/Al TS material has favorable conductivity when compared to wrought Al and cold worked Mo.

4.6 Reactivity measurement by SECM

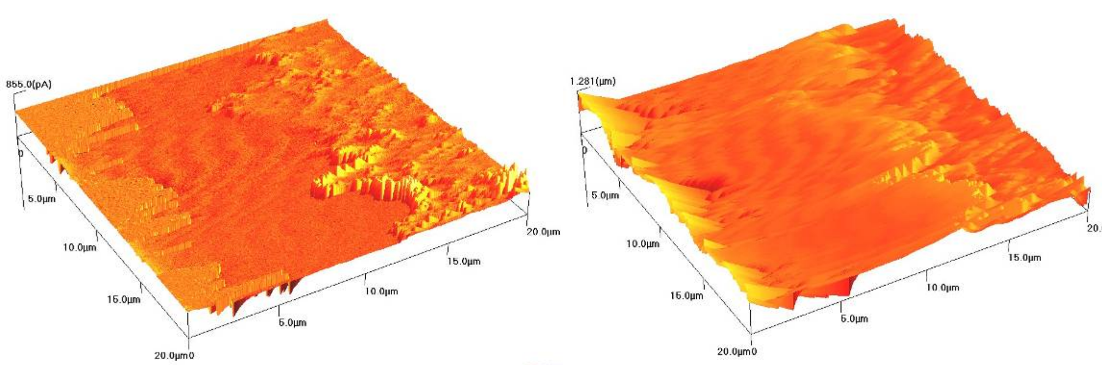

Fig.10. (a) Current (in pA) on the scanned surface at 1V bias potential, (b) Surface topology.

One of the most important characteristics of the metallic BPP is its corrosion resistance. The chemical reactivity of the manufactured bi-metallic plate was characterized by Scanning Electrochemical Microscope (SECM). SECM allows mapping of in-situ topography of the surfaces that are immersed into an electrolyte as well as visualization of spatially confined variations in chemical reactivity (in other words corrosion current under applied bias voltage). Fig. 10 shows the current and topography of the bimetallic plate under acidic solution (HCl, pH6) at a bias potential of 1V. As can be seen from these figures, there was no sharp increase in the current or dissolution of the molybdenum layer indicating an over all good corrosion resistance.

5.0 Current Challenges, Future Work and Conclusion

Progress has been made towards the established manufacturing goal. This exploratory project has demonstrated that 3DP and TS technology can be combined successfully in the creation of metallic parts that meet the requirements of PEMFC BPP. However, further analysis and engineering challenges remain before the next set of developments, illuminated by this initial work, can be pursued.

5.1 Refinement of Thermal Spray Characterization

Further management and characterization of the thermal conditions during the metal deposition phase need to be done in order to provide more accurate "starting point" parameters. By use of the Tecnar plume characterization sensor suite in combination with non-contact coordinated measuring machines (CMM), real time data can be used to quantify thermal conditions and deposition rates and their affect on the dimensional accuracy of the part. Of most importance will be the establishment of optimal substrate temperature ranges.

5.2 Wider Range of Pattern Designs And Guidelines

Similar to any other form of tool design, this process dictates particular feature requirements. More precise guidelines for pattern design will be established. Multiple-stage-sacrificial-pattern-generated-tools will also investigated to create TS deposited parts with increasingly complex features. An interesting point to note is that the geometric freedom offered by the 3DP segment of this work allows for plate geometry that might be able to incorporate constant pressure yielding, spring like features into the monolithic plate design. Also, integration of PEMFC geometry into other vehicular components, such as airfoils is a possibility, as is seen in figure 11.

5.3 Further Improvement of Surface Finish and Inter-Channel Geometry

Of key importance to bi-polar plate performance is electrical conductivity, and a larger contributor to overall conductivity is contact area. If asperities on the deposited parts can be reduced an advance will have been made. The most direct way of improving the surface smoothness of the plate is by improved surface smoothness of pattern (without manual finishing steps). Using permanent, metal patterns to create the flow plates will most probably improve this feature. For prototyping efforts it is important to recognize that consistency of plate conductivity is more important than overall conductivity.

While smooth surfaces are advantageous for good cell conductivity, the ability to create complex specifically textured surfaces on the inside of gas flow channels will be investigated for potential increases gas pressure performance by means of favorable gas-membrane interactions. Additive manufacturing, independent of geometric complexity, has the potential to create inter-channel gas flow designs at no penalty in time or cost.

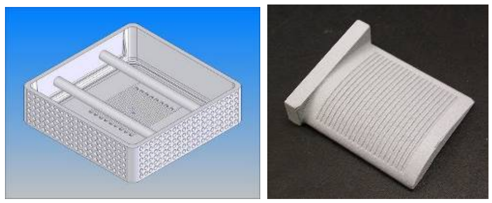

Fig.11. CAD rendering of a 3DP mold for a TS deposited metal forming tool is seen on the left. An example of a complex air foil gas flow component possible with this method.

5.4 Further Material Investigation

While the use of molybdenum is a novel part of this work, and holds promise as a successful material choice, more tests with a variety of metallic and non-metallic, TS deposited materials need to be investigated. The breadth of material choices that can be used in thermal spray opens an interesting area of investigation. Thorough mechanical testing of fabricated parts will also be conducted.

5.5 Permanent Tools

Permanent, H13 tool steel patterns have already been created with an RP investment casting method. These tool will be used to create gas flow plates of the same composition as those made with sacrificial RP pattern, hopefully yielding a process that can be extended into longer run manufacturing. Instead of sacrificing a metal pattern, coatings and precise control of thermal conditions will be used to remove the deposited metal plated from the permanent pattern. Direct metal 3DP, TS deposition tooling or the investment cast tool created already might be extended to limited-run forming tool usage.

The use of TS and RP to create permanent BPP tooling will also be explored. Building on work done at the RML with Ford Motor Company, and elsewhere [18], metal composite tooling will be created with the use of 3DP molds. A mold for a TS deposited tool is seen in Figure 11. This effort might lead to the simplification of the transition to high volume manufacturing, by utilizing the same equipment set involved in prototyping.

5.6 Conclusion

The deployment and global adoption of hydrogen as a renewable energy source is a complex problem that will require utilization of the most current engineering tools. By reducing the limitation of high prototyping and tooling costs, and utilizing geometric-complexity independent, additive methods, progress can be made towards this goal. As the popular slogan goes; “Yesterday’s tools can not solve tomorrow’s challenges.”

This exploratory project has shown that the combination of TS and 3DP technologies can be combined successfully in the creation of metallic parts that meet the requirements of PEMFC BPP. With minimal equipment, material and processing costs a wide variety of BPP and other PEMFC parts can be rapidly produced for functional prototyping. The new methods described in this paper also have potential to be directly transitioned into high volume manufacturing. Further research is required to bring these methods to a state of commercial readiness.

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