MEA manufacturing using an additive manufacturing process to deposit a catalyst pattern in an MEA and its impact on cost reduction.

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Abstract

The manufacturing of a fuel cell Membrane Electrode Assembly (MEA) is a significant cost driver in polymer-electrolyte membrane (PEM) fuel cell technologies, primarily due to the inclusion of expensive materials in the catalyst layer. The selective deposition of a catalyst on the MEA of a fuel cell can drastically reduce the costs depending upon the catalyst, method used for deposition, and production volume. In this paper, testing and analysis of a novel catalyst iridium oxide is discussed. The performance of the catalyst will be compared with the conventional catalysts which will give us an estimate of its effectiveness however, in this paper, only its feasibility in terms of cost is discussed.

1. Introduction

The proton exchange membrane fuel cells (PEMFCs) have been regarded as systems that have a lot of potential applications in transportation, generation of power, and portable electronic devices such as mobile cell, laptops and vehicles, resulting from their high energy-conversion efficiency, low temperature of operation, and environmental benefits [Li 2009].

The components of a PEM fuel cell stack are listed down below:

- Bipolar Plates
- MEA (Membrane Electrode Assembly) containing the proton exchange membrane (Nafion ®) and the GDL (Gas Diffusion Layers) with a catalyst layer coated on it.

The MEA (Membrane Electrode Assembly) is the most important component of a fuel cell as it is the power generating unit. It is also the most expensive component of a fuel cell, primarily due the catalyst layers coated onto the GDL of the MEA.

The high cost of the MEA is primarily due to two reasons: The expensive catalyst and the expensive and time consuming manufacturing methods. There are several ways which can effectively reduce the cost of the MEA. The different ways to do that are:

- Low cost methods of manufacturing
- Reduce the catalyst loading in a way that the performance is not optimized. This can be achieved by optimizing a uniform distribution of the catalyst on the GDL so that a low loading can give equivalent or a better performance
- Use a non-noble or a low cost catalyst.

Replacing platinum has been a goal for the PEM fuel cell industry since a long time because of the price and scarcity of the element on earth. The high loading required remains one of the major drawbacks which are responsible for the inability to manufacture fuel cells on a mass manufacturing basis [Qiao 2009]. Reducing the loading of the platinum requires the particles to be of smaller size, in the range of a few nanometers. The smaller size in turn ensures the higher surface area utilization. However, to make the size of the particles smaller, it requires energy intensive processes such as ball milling and time consuming electrochemical deposition processes. This results in increase in the processing cost which will lead to the increase in cost of fuel cells.

1.1 Alternatives to Platinum

Analysis of literature shows, that in last few years, tremendous progress in improvements of activity and stability of non-platinum cathode catalysts was achieved [Serov 2009]. Several non-platinum materials such as transition metal oxides such as Fe and Co based oxides [Subramanian 2006, Lefèvre 2008], pyrolyzed macrocyclic compounds [Zhang 2006], Ru- based catalysts [Súarez-Alćantara 2006, Pérez 2009]. In spite of this progress, very few catalysts showing real promise to comparable Pt in the real H2/ O2 (air) fuel cell test have been reported so far.

The reason for people looking for alternatives to platinum other than the cost, is that platinum is not tolerant to even trace amounts of CO in a fuel cell and due to the pollution, we have a lot of CO in our atmosphere. The performance of the fuel cell with trace amounts of CO in it decreases substantially. The maximum power density for the heat treated 30% NiWC/KC was 8.2 mW/cm^2 which is only 5-7% of the 20% Pt/C which is the catalyst used in most of the cases [Serov 2009]. Maximum performance in terms of power density from Tungsten carbide W$_2$N/C (18wt%) with a loading of 0.644 mg/cm^2 was 16.02mW/cm^2 as investigated by Zhong et al[Zhong 2007]. Mo as well as its oxides is not catalytically useful for oxygen reduction [Zhong 2006].

The rest of the transition metal nitrides and carbides have the same issue of underperformance. This issue can be addressed with higher loading of catalyst. However, the higher loading of catalysts will upset the formation of triple phase boundaries which will in turn result in less number of active catalyst sites. This leads to a conclusion that, using a large amount of inexpensive catalyst would not give the performance equivalent to platinum. It would rather hinder the performance and would degrade faster than platinum. This would prove to be a costly affair. Hence, we have to look for a catalyst which can give equivalent performance or more as compared to platinum which would not require heavy loading either. A General Motors review states that a power density of 0.7 W/cm^2 at an electrical efficiency of 58% is necessary for a PEM fuel cell [Gasteiger 2005]. Using lower loading of Pt can achieve the goal of efficiency and low cost, however, low loading of Pt results in degradation of the performance of the fuel cell in a faster way. Hence, there is a limit to even how low you can load the Pt as a catalyst in the membrane electrode assembly of a fuel cell.

Of all the technological challenges encountered by the PEM fuel cells, one of the most important is creating an anode catalyst which can tolerate up to 50 ppm of CO [Wee 2007]. IrOx has been considered as CO tolerant catalyst. IrOx has been proposed as good promoters in Pt/C

catalyst because of their excellent oxygen evolution, high activity on hydrogen oxidation, strong affinity for OH or O species and stability in acidic media. [Rasten 2003]. There have been various experiments carried out during the last decade to test and verify the catalytic activity of IrOx for water electrolysis using the proton exchange membrane as the solid electrolyte. Generation of $1A/cm^2$ at 1.51V has been reported using the IrOx as the anode catalyst for PEM water electrolysis [Labou 2008].

Other than looking for alternative catalysts, to reduce the costs further, we have to look for manufacturing methods which are better than the conventional ones from the point of view of cost, simplicity and efficiency. This paper discusses some of the conventional process used for manufacturing the MEA as against the method we have used to fabricate the MEA. There is a need today to innovate new low cost methods of MEA fabrication from the point of view of the properties desired in the MEA for high efficiency operation.

There are three factors which influence the catalyst utilization [Yang 2010]
1. Activity of the catalyst particles
2. Proton transport resistance in the catalyst layers
3. Formation of Triple Phase Boundaries(TBP's)

Now, factor 1 depends on the size of the catalyst particles. The smaller the particle size, the more is the catalytic activity. However, if the size is too small, there is a chance of the particles leaking into the acidic media at the boundary between the GDL and the membrane. Factor 2 is depending on the manufacturing process. We can make the MEA in a way that the thickness of the catalyst layer is not too high or too small. It should be between the optimum levels of thickness i.e. between 15-30 μm. If only the thickness is between these values, the proton is transported effectively and faster from one side of the membrane to the other. This results in high power output. Factor 3 depends on the concentrations of H_2 and O_2 molecules in the catalyst layers, and the diffusivity of H_2 and O_2 molecules in the catalyst layers, and is thus controlled by the H_2 and O_2 flow rates, the structure of catalyst layer, and the porosity of catalyst layer. Thus it is evident that, the catalyst activity depends on the manufacturing technique.

2. Conventional techniques to fabricate the MEA

The conventional techniques to manufacture MEA are:

2.1 Hand painting/Brush painting

The Hand Painting method is one of the most prominent and widely used methods for applying the catalyst layer on to the Gas Diffusion Layer.

Hand Painting Method

Gas Diffusion Layer → Paint (5 min) → Dry (5 min) → Mass (1 min) → Electrode

Repeat until desired loading
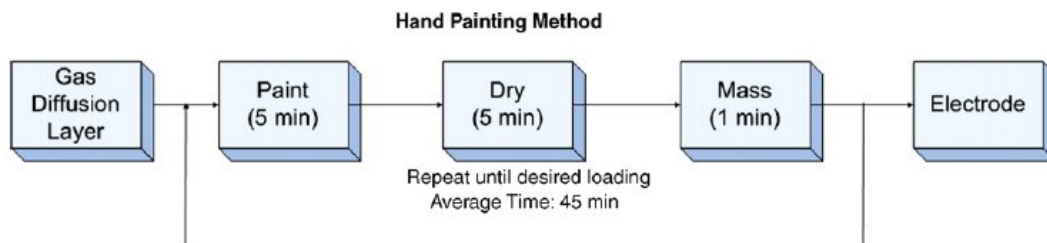Average Time: 45 min

Figure 1 Time duration for the brush painting method [Taylor 2007]

In Brush Painting, we have to iterate the procedure of painting, drying and massing a number of times before the final GDE is obtained for the MEA manufacturing process. This increases cost and cannot be scaled to high volumes. Also, there are inherent problems in the process itself. The uniformity with which the catalyst is distributed is not easily controlled. It will also depend person to person whoever is responsible for application of the catalyst. Hence it is not a process in which the components are easily reproducible [Taylor 2007].

2.2 Spray painting

Spray painting is an evolution of the brush painting procedure. It is an automated assembly and can easily produce similar components in high volumes. But it hasn't been very successful in reducing the loading of the catalysts. Also, periodic clogging due to the catalyst particles takes place thus requiring frequent maintenance [Taylor 2007].

2.3 Sputter deposition methods

Sputter deposition is widely used for integrated circuit manufacturing and has been investigated as a preparation method for PEMFC electrocatalysts for more than a decade. The sputter coating technique has been widely investigated for its success of achieving ultra low loadings of the catalyst and its scalability too. This method could allow for large scale production, however, the initial investment is still substantial due to costs associated with clean rooms, Pt targets, and ultra high vacuum equipment. Also, there is a high possibility of wastage of the catalyst if in case the sputter gun is not directed correctly to its target. The wastage might also be due to reflection from the target in case the catalyst particles fail to adhere to the GDL. In addition, the Pt deposited is often unsupported and the electrolyte cannot be deposited simultaneously with the Pt limiting the catalyst layer to only two dimensions [Taylor 2007].
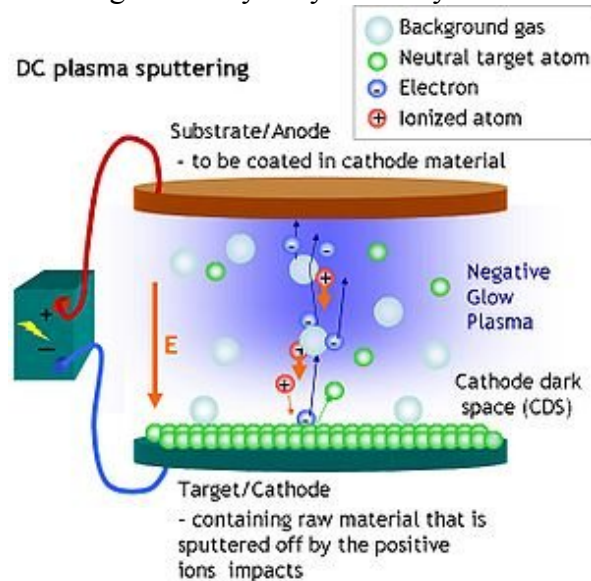

Figure 2 Sputter deposition method [Molhave 2006]

As seen from the figure, it requires sophisticated equipment which is expensive and often prone to breakdowns and hence further increase in costs. The high efficiency of the equipment is thus easily offset by the costs.

3. Novel methods of printing

Nowadays, with an aim of reducing the cost, different techniques of manufacturing are sought after. Some of the new techniques are additive manufacturing techniques such as inkjet printing, electrospraying and direct-write deposition techniques.

3.1 Inkjet printing

Inkjet printing cannot be considered as a conventional process for MEA fabrication. However, recently there has been a lot of interest with respect to the inkjet printing technology for manufacturing the catalyst layers onto the MEA's. Towne et al. [Towne 2007] studied the possibility of manufacturing the GDE (Gas Diffusion Electrode) for PEM fuel cell MEA using off the shelf inkjet printing systems. Although they successfully demonstrated the high capabilities of the process, there are few issues with the process. The drop size, jetting velocity, print head speed, number of nozzles, distance between nozzles, and platen speed cannot be manually controlled. Moreover, the nozzles of the print heads are very small in size, in the range of 10-20 μm. This leads to coagulation of droplets in the print heads. So, to avoid these problems, the catalyst powder has to be ground fine using high cost equipments such as ball mills etc. The catalyst powder size must be around 100-500 nm for the ink droplets to not coagulate in the print heads. This in turn increases the cost of the catalyst powders.

3.2 Electrospraying process

Electrospraying is a process where the flow of the ink onto the GDL occurs because of the light push from the syringe piston and the electric field which exists between an anode and a cathode. The voltage of the electric field depends on the optimum conditions necessary for the deposition to take place. The advantage of the method is its simple technique and low cost equipment. However, it is a relatively slower process thereby offsetting the costs saved with inexpensive equipment. In this paper, the reviewing and analyzing this method with a new process called as electro-write deposition process to with regards to total cost for depositing an optimum quantity of ink is carried out.

3.3 The electro-write process

The electro-write deposition process combines electrospraying and direct write processes into one single process. In an Electrospraying process, the voltage induced by the two electrodes and the electric field acts as a pulling force for the ink to deposit onto the GDL. In the electro-write process, the stepper motor is pushing the syringe as well as the electric field is acting as the pulling force. Hence there is a better scope in this process for the formation of triple phase boundaries.

As stated above the conventional methods have few disadvantages over the electro-write deposition method. However, our present effort is to merely check the feasibility of the electro-write deposition process for manufacturing of the MEA and to check its scalability and its advantages over the conventional processes.

- The electro-write deposition process allows the preparation of precise catalyst content and thickness.
- It can deposit much smaller catalyst particles which increases the Pt surface area, activity, and utilization.
- The method facilitates the fabrication of a nano-scale Pt layer with uniform distribution
- The preparation process is simple and easy to scale-up
- The process allows Pt deposition onto various PEMFC substrates such as GDL, membrane and NCL (or Nafion-bonded NCL).

In this paper, the feasibility and cost analysis of the electrospray process is covered. The MEA manufacturing process consists of three parts:

1. Catalyst Ink preparation
2. Catalyst spraying
3. Iso-static hot pressing of the membrane between the two GDE (Gas Diffusion Electrode)'s.

The only process which is analyzed by comparing two different manufacturing methods is the catalyst spraying process. In this process, the spraying of the catalyst ink on to the GDL takes place to manufacture the GDE (Gas diffusion Electrode). Because of the costs associated with it and the quality of the final product, this component becomes the most important component of the fuel cell. The paper discusses the cost model of the MEA manufacturing process by considering electro-deposition as the base process. The important factors for preparing the GDE by the catalyst spraying are the wettability, ability to soak in the catalyst ink without letting it evaporate which results in the formation of the triple phase boundaries and processing time. The processing time is the one which determines the costs associated with the MEA. However, other factors determine the performance of the fuel cell. Hence everything has to be taken everything into consideration while choosing the right manufacturing process.

4. The Experiment

The Electrospraying process was taken into consideration initially for the manufacture of GDE. A 5cm*5cm Toray cloth was the GDL for the process. The Iridium oxide catalyst content was 25 mg/10ml of ink. Hence, for achieving a loading of 0.4mg/cm^2 which is considered as a standard loading, we prepared only 4 ml of ink. To achieve uniform dispersion, it was stirred additionally for 1 hour before it was loaded in the syringe.

The optimum parameters for the Electrospraying process were fixed by experimentation. In this, one parameter which was the droplet ejaculation rate was fixed and the other two parameters which are the voltage and the distance of the needle from the GDL were varied. The droplet ejaculation rate was also found out by a similar method in which the other two factors were fixed and only the droplet ejaculation rate was varied to find out the optimum rate at which the droplet is absorbed into the GDL without formation of the drop on the surface of the GDL.

In this way, we found out that following parameters are the best for the Electrospraying process to manufacture a GDE.

Voltage	Droplet Ejaculation Rate	Distance from the GDL
3	75-80 µL/min	0.25 cm

After this task, the Electrospraying apparatus is mounted on a XYZ table and automatic Electrospraying of the catalyst ink onto the GDL is carried out. From this study, the optimum traversing speed for the Electrospraying process was found out to be 2 in/min for the entire quantity of ink to spread uniformly on the surface of the GDL. This optimum speed is found on the basis of visual inspection of non-formation of any droplet on the surface of the GDL.
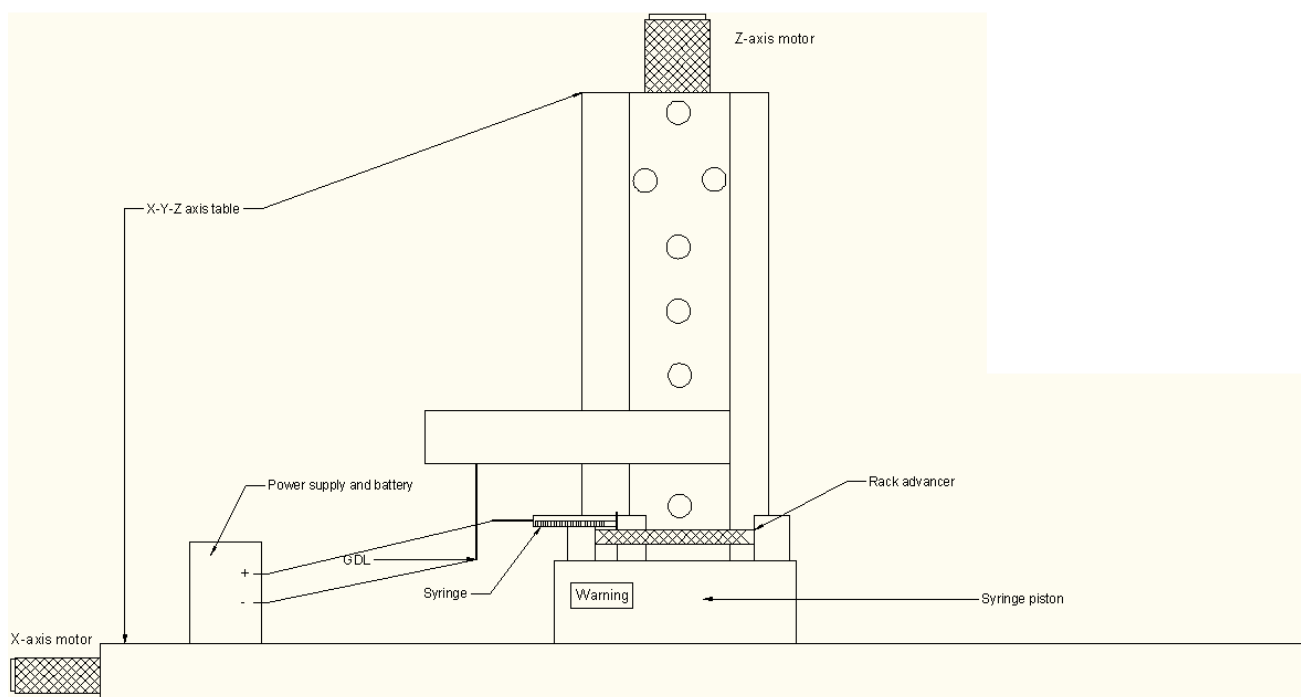

Figure 3 Electrospray apparatus

The pattern in which the ink is sprayed on the GDL is shown below. The spraying was started from the left bottom end of the GDL and then the progress is as shown in the figure. Totally, it took 25 minutes for the entire GDL to be sprayed with the optimum amount of ink which is 4 ml.
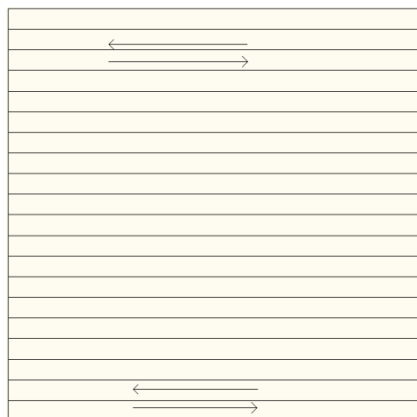
Figure 4 Pattern used for depositing ink in the Electrospraying process

After making the GDE, it was analyzed under a microscope equipped with a 40x zoom. Samples were cut from different parts of the manufactured GDE and analyzed to know the regularity of the catalyst onto the surface of the GDE. Below are some of the pictures of the GDE. Next are some of the pictures from different parts of the manufactured GDE.
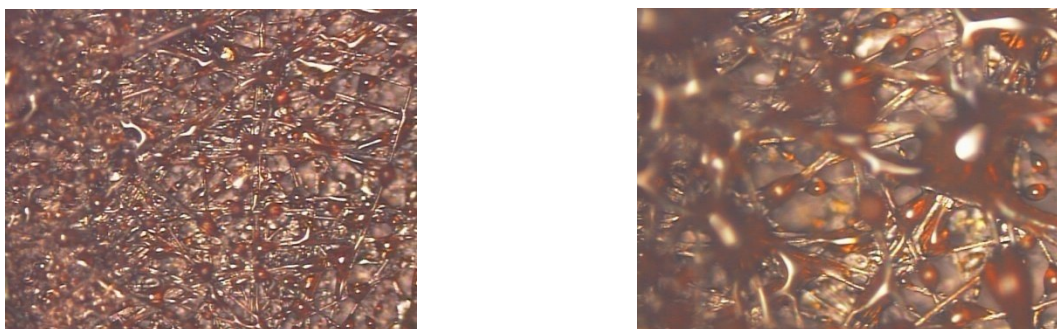
Figure 5 (Left side, 20x zoom) The image shows a sample with droplets regularly distributed. The brown color signifies the presence of the catalyst. (Right side, 40x zoom) The catalyst ink not only present on the upper layers but also on the layers below.

5. Catalyst Ink preparation process

Before describing the cost model, it is important to describe the catalyst ink preparation process. Catalyst ink, containing the catalyst, an electrolyte (ionomer) and certain solvents, is a liquid precursor of the catalyst layer. When preparing the ink, the contents of each component are the first consideration, followed by its uniformity and viscosity [Zhang 2008]. A dispersing agent was used to control the uniform of the dispersion. The components which were chosen to be the part of the ink composition were Isopropyl alcohol, Nafion liquid (5 wt %), Iridium oxide catalyst particles. The quantities of each of the component were taken after a lot of careful study and to achieve the necessary viscosity and the surface tension. The target parameters were:

Viscosity: 1-5 cP

Surface Tension: 30-35 mN/m.

6. Cost model for the Electrospraying process

The cost model for the electrospraying process was calculated considering the cost of the process per liter of catalyst ink. The catalyst loading considered for the MEA is 0.2 mg/cm^2, a standard loading according to previous literature review.

As we know that every process has certain wastage associated with it, we considered a wastage rate of 10% for each of the process.

Components required for making the catalyst ink

Ingredient	for 10 ml	for 1 liter	Cost ($)
Nafion liquid	1ml	100 ml	105
Catalyst particles	10mg	1 g	110
Isopropyl alcohol	8ml	800 ml	5
Dispersing agent	1 ml	100 ml	5
Total cost			225
Final Cost			**247.5**

Final cost = Total cost*110% (10% wastage considered)

Catalyst Ink preparation cost

Description	Elaboration	Cost/Quantity
No of stirrers		2
Stirrer cost /liter	170/240	~0.71
power consumed		1
1 liter beaker cost for 2 units		18
Total operational cost		**19.71**

MEA component costs

Number of MEA's which can be prepared with a loading of 0.2 mg/cm^2 using 1 liter catalyst ink = 100

Component	Units	Cost
GDL	100	235
MEA	100	750
Total material cost		985
Final Cost		**1083.5**

Final cost = Total cost*110% (10% wastage considered)

The electrospraying apparatus cost

Activity		Time/Cost
Catalyst spraying time with electrospray apparatus		30 minutes
Considering there are 10 machines in the shop		300 minutes
Setup time for 1 run		10 minutes
Labor cost ($20/hr, 4 employees)		480
Equipment cost for 10 syringe dispensers	3200/240	13.33
Battery and power supply cost for 10 dispensers	2220/240	9.25
Syringe costs for 1 liter ink	20 syringes/liter	10
Total operational cost		512.58
Final cost		**563.84**

Final cost = Total cost*110% (10% wastage considered)

Hot pressing cost

The best parameters identified for the hot pressing post processing are 1000 psi, 100°C and 2 minutes [Therdthianwong 2007]. Also, the equipment depreciation has to be taken into consideration while computing the process cost. The life time of the hot pressing apparatus is considered to be 7 years.

Hot pressing apparatus cost = 2 *25000/ (7*240).

Here the life of the apparatus is considered as 7 years. The number of working days is considered as 240 in a year. There are 2 machines which are required for the process.

Activity	Cost
Hot pressing apparatus cost	29.7619
Labor cost ($20/hr,1 employee)	100
Total operational cost	129.76
Final cost	**129.76**

Since the wastage cost has been considered in all the earlier stages, it is not considered in the hot pressing operation. The addition of all the final costs is stated as below:

Operation	Cost
Components for catalyst ink	247.5
Catalyst ink preparation	19.71
MEA components	1083.5
Electrospraying	563.84
Hot pressing	129.76
Total cost	2044.31

Thus the total cost of preparation of 100 MEA's comes out to be $2044.31. Hence the cost per piece is $20.44, which is considerably cheap as compared to the stock selling MEA's. However, it is understood from the overall study that the cost depends primarily on the process of GDE preparation since rest of the process are common to any other manufacturing method. Hence, if the process of GDE preparation is fastened or made less labor intensive, huge savings of cost can be achieved.

Conclusion

One of the aims in this study was to demonstrate the feasibility and simplicity of the electrospray deposition process. However, the possibility of cost saving using an additive manufacturing process was the main goal. There are various advantages in the process such as precise deposition of costly catalyst, ability to deposit the catalyst in various patterns. The other advantages in this process are the reproducibility of the catalyst layers in the MEA's. As it is a precise deposition process, we can achieve thin films which can give high power density using low loadings of the catalyst. It is a very agile and flexible method of manufacturing and at a low cost. The cost analysis shows that there is a further scope for cost reduction using faster additive manufacturing methods and that the lesser the time involved in the catalyst spraying process, the lesser is the cost. Further and deeper research needs to be carried out with respect to MEA manufacturing using additive processes. However, there are certainly questions about its scalability which need some thought input.

Acknowledgements

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