

State of Research Developments in Direct Methanol Fuel cell

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ABSTRACT—Purpose – This paper reports the extent and trends of theoretical developments in the Direct Methanol Fuel Cell (DMFC) literature published between the years 1995 to 2016 and provide research gaps that can be bridged by the future research.

Design/methodology/approach – This content analysis based systematic literature review analyzes 56 research articles published in refereed international journals in the discipline of Fuel Cells. These articles are categorized into five article types namely, parameters, Flow field design, water management, Methanol crossover, and Liquid water transport in the cathode following the framework. Analyses are carried out in order to unearth important trends.

Findings –Fuel Cell development is making adequate progress in the DMFC discipline. DMFC discipline has borrowed theories from other disciplines such as PEMFC, AFC, PAFC, MCFC, and SOFC. The most impactful constructs include environmental technology Flow field design, water management, Methanol crossover, and Liquid water transport in the cathode. The paper also identifies the scope for indigenous DMFC.

Research limitations/implications - The findings are based on the research articles published in the Fuel Cell discipline.

Practical implications – The paper highlights predominant theories, frameworks, and constructs that can be utilized by practitioners to improve their understanding of DMFC, their ability to predict future scenarios and solve practical problems.

Originality/value – The Fuel Cell has been used in automobiles and has bright future in our country. So it is hoped that this study will play a significant role in further development DMFC discipline.

Keywords -- DMFC, Flow Channel Design, Water Management, Methanol Crossover

Paper type -- Literature review

I. INTRODUCTION OF FUEL CELL

Fuel cell technology is finding global importance due to less greenhouse gas (GHG)

emissions associated with energy production and influence on climate change. A fuel cell is an electrochemical device that continuously converts chemical energy into electrical energy as long as fuel and oxidant are supplied. Fuel cells therefore have similarities with batteries and engines. With batteries, fuel cells share the electrochemical nature of power generation and with engines, they produce power by consuming fuels. Hydrogen is used as fuel in fuel cells and it generates power and water with zero emission. A difference between a fuel cell and a battery is that a fuel cell generates by-products and the system is required to manage those. A battery also generates some heat but at a much lower rate that usually does not require any additional equipment.

A fuel cell can convert more than 90% of the chemical energy of a fuel into electricity. Typical reactants for fuel cells are hydrogen and oxygen; however neither of them has to be in its pure form. Hydrogen may be present either in a mixture with other gases, such as carbon dioxide, nitrogen and carbon monoxide, or in hydrocarbons such as natural gas and methane, or even in liquid hydrocarbons such as methanol. Ambient air contains enough oxygen to be used as oxidant in fuel cells.

There are different types of fuel cell like Direct Methanol fuel cell (DMFC), Proton Exchange Membrane fuel cell (PEMFC), Alkaline fuel cell (AFC), Phosphoric Acid fuel cell (PAFC), Solid Oxide fuel cell (SOFC).

The DMFC technology is mostly used for small portable power applications like Mobiles, Laptops, Tablets etc., DMFC is having more scope among other fuel cells and that functions similar to PEM fuel cell expect the type of fuel used (fuel for PEMFC is Hydrogen & fuel for DMFC is Methanol).

In this context, following research questions are formed to reveal the state of art and research scope in DMFC.

- What is the extent to which developments, in terms of experimental and mathematical, have taken place within the quantitative DMFC literature?
- How is this trend changing with time and what is its impact on the DMFC literature?

- How can DMFC literature be categorized with respect to theoretical developments i.e. what are the prominent article types with respect to developments within the DMFCs literature?
- Classification of research areas and the extent to which research in fuel cells have taken place?
- Identifying the development of constructs and taxonomies related fuel cells.
- Progress of research (based on citation per year)

These questions are of significant importance as these will clarify the nature of development i.e. experimental and mathematical in the DMFC literature thereby identifying gaps for future research. At the same time, the readers will understand the state of art in the DMFC literature and the level of ideas borrowing from other fuel cell disciplines.

II. CLASSIFICATION OF LITERATURE

A. Developments in terms of Experimental and Mathematical Research

Several researchers identified the paucity of development in the experimental and mathematical discipline since the 1995s (e.g. C. Pu, 1995; J.S. Wainright, 1995; J.T. Wang, 1996; M.K. Ravikumar, 1996). The lack of unifying theory in the experimental and mathematical discipline forces to borrow theories from other disciplines. Liu (2006) argued that the research designs could play a significant role in fuel cell development in the discipline of experimental and mathematical.

Thereafter, researchers engaged themselves with research in the experimental and mathematical discipline and attempted to trace linkages of DMFC with various constructs like Methanol oxidation and oxygen reduction kinetics (A.S. Arico et al., 2000; X. Ren et al., 2000; P. Argyropoulos et al., 1999; J. Nordlund et al., 2004; T. Bewer et al., 2004; J.P. Meyers et al., 2002), Gaseous carbon dioxide in anode (G.Q. Lu et al., 2004; H. Yang et al., 2005; T.S. Zhao et al., 2005; Q. Liao et al., 2007; M.D. Lundin et al., 2007), Liquid water transport in the cathode (M.M. Mench, 2003; G.Q. Lu et al., 2004; K.S. Chen et al., 2005), Methanol crossover (V. Tricoli et al., 2000; S. Hikita et al., 2001; H. Dohle et al., 2002; K. Ramya et al., 2003; R. Jiang et al., 2004 and more), Water management (G.Q. Lu et al., 2005; S.S. Sandhu et al., 2005; F. Liu et al., 2006; W. Liu et al., 2007; M.H. Shi et al., 2007 and more), Flow field design (A.S. Arico et al., 2000; H. Yang et al., 2005). However, no attempt has been made in the past to conduct a best result analysis with regards to performance in the DMFC discipline. The review of literature is fundamental to examine the nature of work published within a discipline and to identify research gaps to be addressed in the future.

A number of frameworks for *experimental and mathematical* in the DMFC discipline have been presented in the past. The details of literature published in the above area are listed in Table I.

III. TABLE I

LITERATURES INCLUDED THE CLASSIFICATION OF APPROACH BY VARIOUS AUTHORS

S. NO	NAME OF THE LITERATURE AUTHOR	MATHEMATICAL APPROACH	EXPERIMENTAL APPROACH
1	Aarne Halme <i>et al.</i> [1] (2016)		*
2	Andrea Calabriso <i>et al.</i> [2] (2015)		*
3	Balaiah Kuppan <i>et al.</i> [3] (2012)		*
4	Chunguang Suo <i>et al.</i> [4] (2014)	*	
5	Chun-Chen Yang <i>et al.</i> [5] (2009)	*	
6	Halim F.A. <i>et al.</i> [6] (2014)	*	
7	Jean Marcel R. Gallo <i>et al.</i> [7] (2012)	*	*
8	Joan M. Ogden <i>et al.</i> [8] (2011)		*
9	Minoru Umeda <i>et al.</i> [9] (1999)	*	
10	Mullai Sudaroli.B <i>et al.</i> [10] (2012)	*	
11	Phuttachart.T <i>et al.</i> [11] (2014)		*
12	Vasco S. Silva <i>et al.</i> [12] (2005)	*	
13	Arico <i>et al.</i> [13] and Ren <i>et al.</i> [14] (2000)		*
14	Argyropoulos <i>et al.</i> [15, 16] (1999)	*	*
15	Nordlund <i>et al.</i> [17] (2004)		*
16	Bewer <i>et al.</i> [18] (2004)	*	*
17	Meyers <i>et al.</i> [19] (2002)	*	
18	Lu <i>et al.</i> [20] (2004)		*
19	Yang <i>et al.</i> [21, 22] (2005)		*
20	Liao <i>et al.</i> [23] (2007)	*	
21	Lundin <i>et al.</i> [24] (2007)		*
22	Mench <i>et al.</i> [25] (2003)	*	
23	Lu <i>et al.</i> [26] (2004)		*
24	Chen <i>et al.</i> [27] (2005)		*
25	Tricoli <i>et al.</i> [28] (2000)	*	
26	Hikita <i>et al.</i> [29] (2001)	*	*
27	Dohle <i>et al.</i> [30] (2002)		*

28	Ramya and Dhathathreyan [31] (2003)	*	*
29	Jiang and Chu [32] (2004)	*	*
30	Kin <i>et al.</i> [33] (2006)		*
31	Han and Liu [34] (2007)	*	*
32	Park <i>et al.</i> [35] (2008)	*	*
33	Kauranen and Skou [36] (1996)	*	
34	Ravikumar and Shukla [37] (1996)		*
35	Cruickshank and Scott [38] (1998)	*	
36	Kuver and Vielstich [39] (1998)	*	
37	Scott <i>et al.</i> [40] (1999)	*	
38	Gurau and Smotkin [41] (2002)	*	
39	Gogel <i>et al.</i> [42] (2004)	*	
40	Du <i>et al.</i> [43] (2007)		*
41	Pu <i>et al.</i> [44] (1995)	*	
42	Wainright <i>et al.</i> [45] (1995)	*	*
43	Wang <i>et al.</i> [46] (1996)	*	
44	Kuver and Kamloth [47] (1998)	*	
45	Lu <i>et al.</i> [48] (2005)	*	
46	Sandhu <i>et al.</i> [49] (2005)	*	*
47	Liu <i>et al.</i> [50] (2006)	*	*
48	Liu and Wang [51] (2007)	*	
49	Shi <i>et al.</i> [52] (2007)	*	
50	Xu and Zhao [53] (2007)	*	
51	Liu <i>et al.</i> [54] (2008)	*	*
52	Arico <i>et al.</i> [55] (2000)		*
53	Yang <i>et al.</i> [56] (2005)		*

From the above table, the best procedure for doing DMFC analyzes are following mathematical approach is primary process and experimental approach is secondary process. Then the optimal result identified by comparison of both approaches.

B. Developments in terms of Parameter, Design and Material Research

A number of frameworks for *parameter, design and material* in the DMFC discipline have been presented in the past. The details of literature published in the above area are listed in Table II.

IV. TABLE II
LITERATURES INCLUDED THE MODIFICATIONS BY VARIOUS AUTHORS

S.N	NAME OF THE LITERATURE AUTHOR	PARAMETER	DESIGN	MATERIAL
1	AarneHalme <i>et al.</i> [1] (2016)	*		
2	Andrea Calabriso <i>et al.</i> [2] (2015)	*	*	
3	BalaiahKuppan <i>et al.</i> [3] (2012)			*
4	ChunguangSuo <i>et al.</i> [4] (2014)		*	
5	Chun-Chen Yang <i>et al.</i> [5] (2009)		*	
6	Halim F.A <i>et al.</i> [6] (2014)			*
7	Jean Marcel R. Gallo <i>et al.</i> [7] (2012)	*		
8	Joan M. Ogden <i>et al.</i> [8] (2011)			*
9	Minoru Umeda <i>et al.</i> [9] (1999)	*	*	
10	MullaiSudaroli.B <i>et al.</i> [10] (2012)	*		*
11	Phuttachart.T <i>et al.</i> [11] (2014)	*		*
12	Vasco S. Silva <i>et al.</i> [12] (2005)			*
13	Arico <i>et al.</i> [13] and Ren <i>et al.</i> [14] (2000)	*		*
14	Argyropoulos <i>et al.</i> [15, 16] (1999)	*	*	*
15	Nordlund <i>et al.</i> [17] (2004)	*		
16	Bewer <i>et al.</i> [18] (2004)		*	
17	Meyers <i>et al.</i> [19] (2002)	*	*	
18	Lu <i>et al.</i> [20] (2004)		*	*
19	Yang <i>et al.</i> [21, 22] (2005)		*	
20	Liao <i>et al.</i> [23] (2007)	*	*	
21	Lundin <i>et al.</i> [24] (2007)	*	*	*
22	Mench <i>et al.</i> [25] (2003)		*	*
23	Lu <i>et al.</i> [26] (2004)		*	*
24	Chen <i>et al.</i> [27] (2005)		*	
25	Tricoli <i>et al.</i> [28] (2000)	*		*
26	Hikita <i>et al.</i> [29] (2001)	*		
27	Dohle <i>et al.</i> [30] (2002)	*		
28	Ramya and Dhathathreyan [31] (2003)		*	
29	Jiang and Chu [32] (2004)	*		

30	Kin <i>et al.</i> [33] (2006)	*		
31	Han and Liu [34] (2007)	*		
32	Park <i>et al.</i> [35] (2008)			*
33	Kauranen and Skou [36] (1996)	*		
34	Ravikumar and Shukla [37] (1996)	*		
35	Cruickshank and Scott [38] (1998)	*		
36	Kuver and Vielstich [39] (1998)	*		*
37	Scott <i>et al.</i> [40] (1999)	*		
38	Gurau and Smotkin [41] (2002)	*		
39	Gogel <i>et al.</i> [42] (2004)	*		
40	Du <i>et al.</i> [43] (2007)	*		
41	Pu <i>et al.</i> [44] (1995)			*
42	Wainright <i>et al.</i> [45] (1995)	*		
43	Wang <i>et al.</i> [46] (1996)	*		*
44	Kuver and Kamloth [47] (1998)			*
45	Lu <i>et al.</i> [48] (2005)			*
46	Sandhu <i>et al.</i> [49] (2005)	*		
47	Liu <i>et al.</i> [50] (2006)		*	
48	Liu and Wang [51] (2007)		*	*
49	Shi <i>et al.</i> [52] (2007)	*		
50	Xu and Zhao [53] (2007)	*	*	
51	Liu <i>et al.</i> [54] (2008)			*
52	Arico <i>et al.</i> [55] (2000)		*	
53	Yang <i>et al.</i> [56] (2005)		*	

From the above table shows that the authors can be changing the parameters involved in DMFC. The flow channel designs and materials have been changed by improving the performance of DMFC. Serpentine flow channel design with porous material is optimal performance of DMFC.

The following table is mentioned the number of citations on the DMFC literature from 1995 to 2016 (Table 3).

**V. TABLE III
CITATIONS**

S.NO	NAME OF THE LITERATURE AUTHOR	# OF CITATIONS
1	AarneHalme <i>et al.</i> [1] (2016)	0

2	Andrea Calabriso <i>et al.</i> [2] (2015)	2
3	BalaiahKuppan <i>et al.</i> [3] (2012)	18
4	ChunguangSuo <i>et al.</i> [4] (2014)	0
5	Chun-Chen Yang <i>et al.</i> [5] (2009)	6
6	Halim F.A. <i>et al.</i> [6] (2014)	1
7	Jean Marcel R. Gallo <i>et al.</i> [7] (2012)	7
8	Joan M. Ogden <i>et al.</i> [8] (2011)	0
9	Minoru Umeda <i>et al.</i> [9] (1999)	440
10	MullaiSudaroli.B <i>et al.</i> [10] (2012)	4
11	Phuttachart.T <i>et al.</i> [11] (2014)	2
12	Vasco S. Silva <i>et al.</i> [12] (2005)	5
13	Arico <i>et al.</i> [13] and Ren <i>et al.</i> [14] (2000)	126
14	Argyropoulos <i>et al.</i> [15, 16] (1999)	761
15	Nordlund <i>et al.</i> [17] (2004)	140
16	Bewer <i>et al.</i> [18] (2004)	188
17	Meyers <i>et al.</i> [19] (2002)	28
18	Lu <i>et al.</i> [20] (2004)	104
19	Yang <i>et al.</i> [21, 22] (2005)	172
20	Liao <i>et al.</i> [23] (2007)	222
21	Lundin <i>et al.</i> [24] (2007)	0
22	Mench <i>et al.</i> [25] (2003)	166
23	Lu <i>et al.</i> [26] (2004)	70
24	Chen <i>et al.</i> [27] (2005)	19
25	Tricoli <i>et al.</i> [28] (2000)	135
26	Hikita <i>et al.</i> [29] (2001)	222
27	Dohle <i>et al.</i> [30] (2002)	135
28	Ramya and Dhathathreyan [31] (2003)	224
29	Jiang and Chu [32] (2004)	105
30	Kin <i>et al.</i> [33] (2006)	145
31	Han and Liu [34] (2007)	122
32	Park <i>et al.</i> [35] (2008)	156
33	Kauranen and Skou [36] (1996)	10
34	Ravikumar and Shukla [37] (1996)	95
35	Cruickshank and Scott [38] (1998)	47
36	Kuver and Vielstich [39] (1998)	108
37	Scott <i>et al.</i> [40] (1999)	369
38	Gurau and Smotkin [41] (2002)	318
39	Gogel <i>et al.</i> [42] (2004)	152
40	Du <i>et al.</i> [43] (2007)	325
41	Pu <i>et al.</i> [44] (1995)	311
42	Wainright <i>et al.</i> [45] (1995)	154
43	Wang <i>et al.</i> [46] (1996)	106
44	Kuver and Kamloth [47] (1998)	180
45	Lu <i>et al.</i> [48] (2005)	1237

46	Sandhu <i>et al.</i> [49] (2005)	278
47	Liu <i>et al.</i> [50] (2006)	90
48	Liu and Wang [51] (2007)	127
49	Shi <i>et al.</i> [52] (2007)	35
50	Xu and Zhao [53] (2007)	142
51	Liu <i>et al.</i> [54] (2008)	63
52	Arico <i>et al.</i> [55] (2000)	8
53	Yang <i>et al.</i> [56] (2005)	86
54	AarneHalme <i>et al.</i> [1] (2016)	71
55	Andrea Calabriso <i>et al.</i> [2] (2015)	147
56	BalaiahKuppan <i>et al.</i> [3] (2012)	166

The nature of research and outcome of the research are discussed in the following sections.

Aarne Halme *et al.* [1] discussed about an alternative concept for DMFC Combined electrolyzer and H₂ PEMFC. This paper presents a novel idea for producing electricity from methanol by using a combination of a PEM fuel cell and a PEM electrolyzer. In some situations the efficiency in making electricity from methanol could be fairly higher. This is owing to the reality that production of hydrogen can be done using very low electrolysis energy on the one case, and in the other way a PEM H₂ fuel cell is more competent than a DMFC. In a PEM electrolyzer the cathode electrode is in a worse potential and no air is participated, which leads to prevent effectively forming of CO from the crossover methanol. According to author experience, the methanol concentration at the electrolyzer anode can be improved without the risk of poisoning the Pt catalyst to much advanced concentrations than in DMFC. From a practical overview, the possibility to work the system using higher methanol concentrations without water dilution is an vital aspect, because it shorten the technical construction.

Andrea Calabriso *et al.* [2] discussed in his study about the CO₂ bubble generation influence on direct methanol fuel cell performance. The presence of gaseous CO₂ produced in the anode channels is the major problem as it delays the free surface of the Gas Diffusion Layer (GDL) decreasing the energetic area and the methanol flux through the permeable media towards the catalyst layer. In this study the manipulation of gas phase fraction on the cell performance and the connection with the operating factors such as methanol-water solution flow rate, air flow rate and current density is explored. The bubbles shows a reduced in mean diameter when the flow rate rose up. The coalescence of bubbles can cause a shrink in generated power in particular when the flow rate is low and the current density is much high. He found that bubbles generation decreases the power also more than 40 %.

Balaiah Kuppan *et al.* [3] discussed the Platinum-supported ordered mesoporous carbon catalysts were prepared employing colloidal platinum decreased by 4 different reducing agents, viz., sodium borohydride, paraformaldehyde, hydrogen and ethylene glycol. Platinum nano particles settled over mesoporous CMK-3 carbon by paraformaldehyde reduction method is better than the other reduction methods. The paraformaldehyde method is more right for the preparation of the high dispersed uniform seized platinum nano particles on the mesoporous support with an average size of the particle resulted in 4 nm. Thus the present paper deals that optimized reduction methods is the appropriate selection of carbon supports can offer major cost savings by lowering the catalyst loading.

ChunguangSuo *et al.* [4] defined about the Design of MEMS-based micro direct methanol fuel cell stack. The silicon-based DMFC stack was proposed as a “flip-flop” style and was manufactured using MEMS technology. Compared to the SS DMFC stack in planar connection, the flip flop connection way can majorly decrease the space of the connection resistance electric and connection. When fed 2M methanol solution and air as oxygen both the stainless steel stack and the silicon stack can power the LED with comparable output of 6.77mW and 6.75mW, correspondingly.

Chun-Chen Yang *et al.* [5] discussed the Preparation of a novel composite membrane and PtRu/Hollow carbon sphere (HCS) anode catalyst for alkaline DMFC. The alkaline composite anionic-exchange membrane is based on PVA/QASP/TAMPFS-PET composite electrolyte was manufactured by a solution casting method. An alkaline DMFC, comprising of the PVA/QASP/TAMPFS-PET composite polymer membrane, was systematically examined and assembled. The maximum power density of alkaline DMFC composing of the PVA/QASP/TAMPFS-PET composite polymer membrane. This membrane used for purpose in alkaline DMFCs.

Halim F.A *et al.* [6] enhanced overview on vapour feed direct methanol fuel cell. DMFC works in two basic behavior which are methanol can be fed in liquid or vapor phase. Due to methanol crossover issue faced by liquid feed DMFC operated at high methanol concentration, vapor feed DMFC is an another way to solve this trouble. Methanol obstacle layer used to increase the mass transfer resistance in the fuel cell hence decrease methanol crossover. Water management layer was added at the cathode to push large amount water backward from cathode to anode to hydrate the membrane for methanol oxidation reaction.

Jean Marcel R. Gallo *et al.* [7] developed Novel mesoporous carbon ceramics composites as electrodes for DMFC. MCC composites for electrodes planned to take the merits of the well ordered mesoporous silica structure and of the graphite which is having high conductivity, is presented for the first time. This behavior can be revealed by the more difficult diffusion of the liquid combustible if evaluated with the gas oxidant. The fuel can diffuse enhanced in the organized porous MCC structure than in the Vulcan XC-72R structure.

Joan M. Ogden *et al.* [8] discussed the study about a comparison of methanol, hydrogen and gasoline as fuels for fuel cell vehicles: implication for infrastructure development and vehicle design. Hydrogen can be stored directly or formed onboard the vehicle by reforming methanol. The vehicle design is easy with direct hydrogen storage. But it needs developing a more intricate refueling infrastructure. In this study, we present modeling results comparing or evaluating three leading choice for fuel storage on board fuel cell vehicles: (a) on board partial oxidation (POX) of hydrocarbon fuels derived from crude oil, (b) compressed gas hydrogen storage on board steam reforming of methanol (c) Considering both vehicle and infrastructure issues. Feasible fuel a strategy tends to the commercialization of fuel cell vehicles are formalized. Hydrogen is favor fuel for fuel cell vehicles, for reasons of cost, vehicle design and efficiency.

Minoru Umeda *et al.* [9] discussed about novel Pt-C and Pt-Ru-C electrodes were made by a co-sputtering technique to increase the methanol oxidation reaction. An O₂-enhancing methanol oxidation at the Pt_{0.56}C_{0.44}, which never occurs for the Pt, was obtained and compared to that for the Pt_{0.61}Ru_{0.34}C_{0.05}. The results exhibited that the addition of Ru to the Pt-C can improve the methanol oxidation current and tends to a negative shift in the onset potential (*E*_{onset}) in mutually the N₂ and O₂ atmospheres. Based on these results showed, the O₂-enhancing methanol oxidation with a negative *E*_{onset} is obviously achieved by the Pt-Ru-C sputtered electrode. This also points out the fact that the O₂-increasing methanol oxidation takes place under these situations. The DMFC power generation performance was then reviewed by changing the counter-electrode reactant from H₂ to O₂. By contrasting the results of the DMFC power generation performance and methanol oxidation voltammogram, the degraded DMFC power generation was resolved due to methanol crossover.

Mullai Sudaroli.B *et al.* [10] explained about Heat and Mass Transfer Characteristics of Direct Methanol Fuel Cell. Temperature distribution and Methanol in the anode side are expected. Double channel flow field is used to explore the methanol

distribution and its consequence on cell performance. Water crossovers and Methanol in the cell are the main controlling factors which control the cell performance. The model is also used to calculate the methanol crossover effect on Cell performance and the Fuel Utilization Efficiency (FUE). The cell efficiency had increased range from 7 to 13% with decreasing methanol concentration of 1 to 0.25 M. The cell efficiency also increases from 7 to 13% due to high FUE.

Phuttachart.T *et al.* [11] fabricates bipolar plates for direct methanol fuel cells (DMFC) to increase creep behavior and compressive strength. The composites consist of poly methyl methacrylate (PMMA), carbon black (CB) and polyurethane (PU) and were manufactured via bulk polymerization in a casting process. The composites were prepared in various weight ratios PMMA/PU/CB. The potential of using PMMA/PU/CB composites as original polymeric bipolar plates for DMFCs. PMMA/PU/CB composites were fabricated by bulk polymerization via casting process were victorious in further improving mechanical properties of the composites such as brittleness, creep behavior and compression strength. To know more characters of composites as a function of bipolar plate role, in-situ and ex-situ creep behavior tests should be examined as future work.

Vasco S. Silva *et al.* [12] examined the Membranes for direct methanol fuel cell applications and an outline of the research development regarding this DMFC module. Specific efforts are given over to research feature related with the characterization, membrane preparation, DMFC tests and modeling. Membranes with increased relation between barrier and electrolyte properties were made in comparison with that of Nafion.

Methanol oxidation and oxygen reduction kinetics

Aricoet *et al.* [13] and Ren *et al.* [14] found that thinner support layers and even unsupported catalyst lead to better performances. The authors also found that smaller particles and higher surface areas are beneficial for the process. Contrarily, the atomic ratio between platinum and ruthenium seems to have a lower influence. For higher operating temperatures, the optimal performance has been found with atomic ratios in the region of 1:1. For lower temperatures a higher relative platinum content seems to be beneficial.

Argyropoulos *et al.* [15, 16] were perhaps with the first to study the two-phase flow pattern in the anode of a DMFC. They used acrylic cells and a high speed video camera for visually investigate the carbon dioxide gas evolution process inside an operating DMFC environment. The effect of operating conditions on the gas management using different gas

diffusion layers (carbon cloth and carbon paper), flow channel designs, cell sizes and exhaust manifold configurations was studied. The visualization studies showed that carbon paper has a relatively low ability to gas removal. Increasing the methanol solution inlet flow rate was beneficial for gas removal. Increasing the current density leads to a higher gas production and in the formation of gas slugs, especially for low flow rates, which can block the channels and lower the cell performance. The new flow channels design, proposed by the authors, based on a heat exchanger concept was more effective for gas management and gave a more uniform flow distribution in the channels than a simple parallel flow channel. This study was, however, undertaken under low cell performance.

Nordlund *et al.* [17] developed a visual DMFC, comprising a transparent anode and a cathode endplate with an incorporated heat exchanger and a picture analysis methodology. They took the data analysis a step further, since they presented a methodology to acquire good visual data and to perform a high-quality and time effective analysis. In particular, they demonstrated how a visual cell in combination with digital video recordings and picture analysis can be used to give precious insight into two-phase flow in the anode of a DMFC.

Bewer *et al.* [18] developed a new method to analysis the bubble generation in an aqueous medium and the interaction of the flow distribution. The method is based on the disintegration of hydrogen peroxide solution to oxygen and water in aqueous media at the presence of a catalyst. By using an appropriate hydrogen peroxide concentration, the gas evolution rate can be set to the same order of magnitude as in real DMFC. This method can simulate the bubble formation in the anode compartment of a DMFC without any electrical current. The current density to be simulated can be adjusted by an appropriate setting of the hydrogen peroxide concentration independently of ohmic losses. As no current conducting parts are needed, the whole cell can be made of a transparent material (perspex) to ensure a complete visibility of the flow. The cell has a simple design in which differently and flow fields can be tested.

As was mentioned and explained before, most of the studies conclude that the reaction can progress according to several mechanisms. However, it is commonly accepted that the more significant reactions are the adsorption of methanol and the oxidation of carbon monoxide adsorbed. So the following reaction mechanism, similar to the one used by **Meyers *et al.* [19]**, was used in the present work.

Gaseous carbon dioxide in anode

Lu *et al.* [20] developed a carefully designed transparent DMFC to visualize, in situ, the bubble flow in the anode of a DMFC. Normally, the transparent cells used in the visualization studies are constructed with a pair of stainless steel plates mated with a polycarbonate plate, forming a window to allow direct observation of flow behaviour. The polycarbonate plate is concave in design while the stainless steel plate had a matching convex pattern to avoid flow leakage. The channels are machined through the stainless steel plate and the surface that contacts with the MEA are coated with Cr (Chromium) and Au (Gold) to minimize the contact resistance. They used two types of MEAs based on Nafion 112 to investigate the effects of the backing pore structure and wettability on cell polarization and two-phase flow dynamics. One employed hydrophobic carbon paper backing material and the other hydrophilic carbon cloth. For the hydrophobic carbon paper they observed that carbon dioxide bubbles nucleate at certain locations and form large and isolated bubble slugs in the channel. For the hydrophilic carbon cloth it was shown that bubbles are formed more uniformly and of smaller size.

Yang *et al.* [21, 22] used a transparent DMFC to investigate experimentally the effect of the single serpentine (SFF) and parallel flow fields (PFF) on the cell performance and on carbon dioxide bubble behaviour. They found that the DMFCs equipped with SFFs leads to better performances than those with PFFs. This investigation was also found that gas bubbles blocked the flow channels in the PFF at high current densities and low methanol solution flow rates. Since fuel cells with PFFs had poor performance they focused their work on studying the effects of various SFF design parameters, including open ratio and channel length, on cell performance.

Liao *et al.* [23] presented a study where a transparent DMFC was constructed to visualize the two-phase flow of aqueous methanol solution and carbon dioxide bubbles by using a high-speed video camera. The dynamic behavior of carbon dioxide gas bubbles including emergence, growth, coalescence and removal was recorded in situ, and polarization curves were obtained to provide an initial understanding of the relationship between the behavior of the cell performance and carbon dioxide gas bubbles. A series of parametric studies, including aqueous methanol solution flow rate, temperature, cell pressure difference and concentration between the anode and the cathode was presented in order to evaluate the effects of these parameters on carbon dioxide gas bubbles behavior and on cell performance. It was observed that gas bubbles first emerge around the corner on the porous diffusion layer and the channel ribs and produced large gas slugs by growth

and coalescence in the channel. The cell performance was improved with increasing aqueous methanol flow rates, feed temperature, feed concentration and pressure gradient between the anode and cathode.

Experimental observation in a small DMFC test cell done by **Lundin et al. [24]** indicated that the rate of bubble formation can be reduced by rising the fuel flow because more liquid is available for the carbon dioxide to dissolve in. The authors also found that potassium hydroxide and lithium hydroxide added to the fuel eliminate in situ carbon dioxide gas formation at low concentrations, because of the consequent greatly increased solubility. They presented a model that explains the rate of carbon dioxide gas formation at the anode of a DMFC including a function of the cells output current, operating pressure, operating temperature, fuel flow rate, and the solubility of carbon dioxide in fuel solution, that is also a strong function of temperature.

Liquid water transport in the cathode

Mench et al. [25] A proper water level of water at the cathode side is necessary to hydrate the polymer membrane, increasing in this way the proton conductivity. However, a too large amount of water in the cathode side leads to water flooding at the pores decreasing the cathode performance. In order to accurately predict critical operation conditions to avoid flooding, visualization of the cathode side is essential to yield fundamental physics behind the flooding occurrence.

Lu et al. [26] developed a carefully designed transparent DMFC to visualize, in situ, the bubble flow in the anode of a DMFC and the cathode flooding. The authors used two types of gas diffusion layers (GDLs), carbon paper and ELAT carbon cloth (formed of carbon cloth type A and treated with Pt on one side). Flow visualization of cathode flooding indicates that more water droplets emerge upon the carbon paper GDL surface than upon the single-side ELAT GDL, due mainly to the higher hydrophobicity of the latter material at elevated temperatures.

Chen et al. [27] presented simplified models which are based on macroscopic force balances and droplet-geometry approximations for predicting the onset of instability leading to elimination of water droplets at the diffusion layer / flow channel interface. They carried out visualization experiments to observe the formation, growth, and removal or instability of the water droplets at the selected interface of a simulated polymer electrolyte fuel cell cathode.

Methanol crossover

Tricoli et al. [28] studied the methanol crossover rate and proton conductivities in two

commercially available, partially fluorinated membranes. The methanol crossover rate was identified by measuring the steady state current at the cathode when methanol was introduced into the anode.

Hikita et al. [29] determined the methanol crossover rates by continuously measuring the concentration of methanol, carbon monoxide and carbon dioxide in the exhaust gas of the cathode. A common method to measure the methanol crossover in a DMFC is the analysis of the CO₂ content of the cathode exhaust. However it is necessary to point out that during the operation of a DMFC a large amount of carbon dioxide is produced in the anodic reaction and some of this can diffuse partially to the cathode side. In this case the amount of carbon dioxide present in the cathode exhaust is a contribution of the carbon dioxide resulting from the methanol crossover oxidation at the cathode side and of the carbon dioxide that passes through the membrane to the cathode side.

Dohle et al. [30] describe a method to separate the two contributions under real DMFC operating conditions and clearly define the amount of carbon dioxide due to methanol oxidation on the cathode side.

Ramya and Dhathathreyan [31] directly measured methanol flux rates across Nafion membranes by an electrochemical method, like cyclic voltammetry and chronoamperometry. The membrane permeability was measured by this technique for various methanol concentrations. The authors found that the permeability of methanol is dependent on the concentration of methanol, the permeability increases with an increase on the methanol concentration.

Jiang and Chu [32] estimated the amount of methanol crossover more accurately with a method of gravimetric determination of barium carbonate to analyse the amount of carbon dioxide. The equivalent current of methanol crossover was calculated from the discharge current of the fuel cell and the amount of dry barium carbonate precipitate collected at the anode and the cathode exhaust. With the method proposed by the authors, the common experimental deviation of measuring methanol crossover caused by carbon dioxide permeation through the polymeric electrolyte membrane can be corrected.

In the work presented by **Kin et al. [33]** the methanol crossover rate of PEM and the efficiency of DMFC were estimated by measuring the current and the transient voltage at the DMFC when methanol was introduced into the anode. The proposed method simply yields an estimate of the methanol crossover rate of PEM and the efficiency of a DMFC and does not require fitting analyses.

Han and Liu [34] determined the methanol crossover rate in a DMFC by measuring the carbon dioxide concentration at the cathode exit in real time, at different inlet methanol concentrations and various operating conditions.

Park *et al.* [35] presented a practical way of characterizing the mass transport phenomena of membrane electrode assemblies (MEAs) through mass balance research in DMFC systems. This method could be used to measure methanol utilization efficiency, the water transport coefficient and the methanol to electricity conversion rate of a MEA in DMFCs. The research on the DMFC MEA design was performed with the aim of reducing methanol and water crossover maintaining high power characteristics. By varying material properties, the critical design parameters were identified for high methanol utilization improving power density through systematic experiments. Due to the impact of methanol crossover on the DMFC performance, its effects have been found out in the literature under various operating conditions, such as, methanol concentration, cathode air pressure, temperature, fuel flow rate, membrane thickness and equivalent weight and catalyst morphology.

In their work, **Kauranen and Skou [36]** studied the influence of the temperature in methanol permeability and found an increase of the crossover rate with temperature.

Ravikumar and Shukla [37] found that in despite of the fact that the increase in temperature increases the methanol crossover, the increase of temperature also leads to an improvement of cell performance, since the ohmic resistance and polarization reduce. The authors also found that the cathode electrode performance is considerably lowered at higher methanol concentration leading to a decrease in cell performance, since higher methanol concentrations result in higher rates of methanol transport through the membrane.

Cruickshank and Scott [38] studied the effects of methanol concentration and oxygen pressure in cell performance and found that higher methanol concentrations lead to a lower cell performance and attributed this to the fuel crossover phenomena. They also found that pressurising the oxygen reduced the methanol crossover leading to higher methanol concentrations.

Kuver and Vielstich [39] studied the effects of methanol concentration, fuel cell temperature and catalyst loading in cell performance. A new catalyst support was found showing a good performance with a smaller noble metal loading.

Scott *et al.* [40] investigated the effect of cell temperature, air cathode pressure and methanol concentration on the power performance on a small-scale DMFC. Higher power densities were achieved at higher temperatures and cathode air pressures. They concluded that the selection of methanol concentration, to obtain maximum power density, depends upon the current density.

Gurau and Smotkin [41] measured the methanol crossover by gas chromatography as a function of temperature, fuel flow rate and methanol concentration.

Gogel *et al.* [42] presented investigations to determine the dependence of the performance of direct methanol fuel cells and the methanol crossover rate on the operating conditions, on the structure of the electrodes and on the noble metal loading. It was shown that performance and methanol permeation depend strongly on cell temperature and cathode air flow. Also, methanol permeation can be condensed significantly by varying the anode structure, but the changed electrode structure also leads to a bit lower power densities. The metal loading was varied at the anode and cathode, affecting the cell performance. Moreover, the differences between supported and unsupported catalysts were compared. They also, discussed the most favorable conditions for the DMFC operation considering the various important factors.

Du *et al.* [43] developed a half-cell consisting of a normal DMFC cathode and a membrane contacting with an electrolyte solution to investigate the effect of methanol crossover on the cathode behaviour. Open circuit potentials, cyclic voltammetry profiles, electrochemical impedance spectroscopy and polarization curves resulting from the oxygen reduction reaction with/without the cause of methanol oxidation reaction, were measured. The steady-state results confirmed that the presence of methanol at the cathode led to an important poisoning effect on the oxygen reduction reaction, especially when the DMFC performs at higher methanol concentrations and discharges at lower potentials.

Pu *et al.* [44] studied a composite electrolyte wherever a film of a methanol resistant protonic conductor, such as a metal hybrid, was sandwiched between proton permeable electronic insulators, such as Nafion. They studied different electrolyte systems where membrane was Nafion 115 with a metal hybrid Pd (Palladium), and the influence of interfaces modification with Pt via a variety of techniques. The results obtained shows that the methanol crossover was smaller than in the Nafion polymer and that the privileged performance was obtained with the system (N/Pt/Pd/Pt/N), where N is the Nafion 115 membrane, treated by palladization and followed by platinization

using electrochemical methods common to the preparation of hydrogen reference electrodes.

Wainright *et al.* [45] studied the conductivity, water content and methanol vapor permeability of the phosphoric acid-doped polybenzimidazole (PBI). Experimental results confirmed the low methanol crossover in a PEM fuel cell employing a doped polybenzimidazole membrane.

Wang *et al.* [46] used the same membranes and studied the methanol crossover and the performance of a DMFC. They observed that the methanol crossover rate increased with a decrease in the water methanol ratio in the anode feed stream. These authors, however, found that methanol crossover increased with increasing current density and methanol crossover decreased when temperature increased. They present in their work, the influence of increasing different operating parameter in the methanol crossover and in the performance of DMFCs using acid doped PBI membranes.

Kuver and Kamloth [47] studied the methanol crossover in substituted and cross linked POP membranes (sulphonate substituted polyoxiphenylenes) by differential electrochemical mass spectrometry. They compared the results with those taken with different commercial membranes and it was found that the superiority of POP membranes was evident, with respect to methanol permeation, and especially attractive since the POP film was only 0.2 to 0.3 mm thick.

Water management

Lu *et al.* [48] reported a novel DMFC design based on a cathode gas-diffusion layer coated with a microporous layer to construct the hydraulic pressure on the cathode side and on a thin membrane, Nafion 112, to promote water back-flow in this difference in hydraulic pressure. Such MEAs exhibit extremely low water flux through the polymer membrane. The importance of the experimental work reported by the authors is the fact that commercially available Nafion membranes and MEA materials were used and the cell performed with ambient air without pressurization.

Sandhu *et al.* [49] developed a mass flux model to predict the fluid phase superficial velocity, methanol and water molar fluxes, and the chemical species (water and methanol) dimensionless concentration profiles in the polymer electrolyte membrane, Nafion 117, of a DMFC. This model can be used to generate the numerical data as a function of different variables, such as the pressure difference across the membrane, temperature, methanol concentration, and position in the membrane.

Liu *et al.* [50] described a new MEA design planned to achieve, simultaneously, low water crossover, low methanol crossover and high power density. They performed general experimental parametric studies to elucidate the effects of material properties, MEA production processes and operating conditions. They observed that the important material properties are structure of the cathode gas diffusion media the membrane thickness. The authors suggest that the key operating factors are the methanol crossover, cathode stoichiometry, cell temperature and current density.

Liu and Wang [51] based on a 3D two-phase model numerically investigated an interfacial liquid coverage model applied at the interface between the cathode supporting layer and flow channel and its effects on the net water transport coefficient distribution in a DMFC were explored under typical operating conditions for portable applications. The authors showed that interfacial liquid coverage has a intense effect on the net water transport coefficient through the membrane by affecting water diffusion and hydraulic permeation.

Shi *et al.* [52] analyzed water transport phenomenon in PEM and the mechanism of occurrence and improvement of a two-phase countercurrent flow with corresponding transport phenomenon in the PEM. A one-dimensional steady state model of heat and mass transfer in porous media system with internal volumetric ohmic heating was developed and simulated numerically to analyze water transport characteristics in the PEM.

Xu and Zhao [53] proposed a measurement method enabling an in situ determination of the water-crossover flux through the membrane in a DMFC. With this method the authors investigated the effects of various design and geometric parameters as well as operating conditions, such as the properties of cathode gas diffusion layer, membrane thickness, cell current density, cell temperature, feed methanol concentration, and oxygen flow rate, etc., on the water crossover through the membrane in a DMFC.

Liu *et al.* [54] experimentally studied various anode diffusion media to reduce the water crossover in a DMFC. A two-phase water transport model was also utilized to theoretically study the effects of those structures on saturation level and water transport in a DMFC anode. It was found that wettability of the anode microporous layer has a dramatic effect on water crossover or on the water transport coefficient through the membrane. Under variable current densities, the MEA with a hydrophobic anode MPL had consistently low values, several times smaller than those with a hydrophilic microporous layer or without an anode microporous layer. A modelling study of anode water transport revealed that the liquid

saturation in the anode was significantly lowered with the increase of the anode MPL contact angle, which was thus identified as a key parameter to minimize water crossover in a DMFC.

Flow field design

Arico *et al.* [55] the interdigitated flow field significantly enhances mass transport and membrane humidification, in a DMFC, allowing higher maximum power outputs compared to the serpentine flow field. The DMFC equipped with serpentine flow field showed however lower methanol crossover, higher fuel utilization and a somewhat large efficiency

at low current densities. When comparing the parallel and serpentine flow field the results.

Yang *et al.* [56] describes the serpentine flow field revealed a better performance than the parallel flow field they focused their studied on the effects of the open ratio and channel length of the serpentine flow field on the cell performance and pressure drop. The studies indicated that the open ratio and flow channel length have important effects on the cell performance and pressure drop. When designing a serpentine flow field, caution has to be taken to ensure an optimal open ratio and flow channel length.

The results of citation per article analysis are presented in below Table 4.

**VI. TABLE IV
CITATIONS PER ARTICLE ANALYSIS**

YEARS	Total no. of articles	MATHEMATICAL APPROACH			EXPERIMENTAL APPROACH			BOTH APPROACHS		
		Total Articles	Total Rating	Mean Rating	Total Articles	Total Rating	Mean Rating	Total Articles	Total Rating	Mean Rating
1995 – 2000	16	12	3680	306.67	7	2968	424	3	1565	521.67
2001 – 2005	19	11	1426	129.64	13	1606	123.54	5	522	104.4
2006 – 2010	12	9	588	65.33	7	490	70	4	355	88.75
2011 - 2016	9	4	12	3	6	29	4.83	0	0	0

The citation of published literature reveals that more number of researches has taken place during 1995 to 2000. A citation per article was high in experimental research during the year 1995 to 2000. The citation per article is decreasing year by year. From the results we could arrives a decision that research focus might shift to PEM fuel cell. The reason may be higher efficiency of PEM fuel cells (40% - 60%). But safety is major issue in the use of PEM fuel cells in applications like Mobiles, Laptops and Tablets etc.,

VII. CONCLUSION

Literature investigation has revealed various approaches involved in a direct methanol fuel cell research.

Direct methanol fuel cell technology has been considered as the next big thing in the field of alternative power sources and has shown signs of potentialities to replace conventional batteries in application in portable electronic devices. However, despite several years of active research, there still exist several drawbacks associated with its basic operation, which are preventing the widespread use of

this potentially promising technology. Present review shows the drawbacks and tried to identify and critically scrutinize the various factors and parameters associated with challenges in DMFC real time user.

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