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MOLECULAR SIEVE AS AN ECONOMICAL ROUTE FOR THE REMOVAL OF HYDROGEN SULFIDE (H_2S) AND TETRAHYDROTHIOPHENE(THT) FROM NATURAL GAS FOR FUEL CELL APPLICATIONS

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The removal of hydrogen sulfide (H_2S) and tetrahydrothiophene (THT) from the natural gas has been studied by employing two kinds of sorbents, i. e., compounds of sulfur-absorbing and sulfur-adsorbing materials. For the removal of sulfur compounds, a system was designed, established and operated for 2000 hours. Determination of the life-cycle assessment and feasibility of material's effectiveness were also carried out. The same experiment was repeated by using a substitute (molecular sieve) and the treated gas coming out of the cleaning system was examined every 4-hour-run time by Dräger gas detector pump (DGDP) for the presence of H_2S and THT. The results were very encouraging for this material. The molecular sieve is found highly economical as a single material with a simple design when compared with the set of sorbents tried earlier. The sulfur-free natural gas is required as feedstock for the production of hydrogen gas which will be consumed by the proton exchange membrane fuel cells for smooth functioning, keeping in view the high efficiency of the system.

Keywords: Sulfur compound removal, Natural gas, Sorbents, Molecular sieve, Hydrogen, Hydrogen sulfide, Tetrahydrothiophene, PEMFC, Mercaptans.

1. Introduction

The unavoidable socio-economic impact of recurring fuel crisis, increasing environmental pollution and the obvious effects of mankind induced climatic changes made the development of clean and renewable energy resources inevitable. Fuel cells are undoubtedly one of the key alternatives to replace conventional energy means as a clean electrical energy source for both transportation and stationary applications. Since 1960, when NASA for the first time used fuel cells in a space shuttle as a backup of electric supply, the research work gained momentum on all types of fuel cells. Over the past two decades Polymer Electrolyte Membrane (PEM) fuel cells emerged as the leading fuel cell type for portable and automotive applications due to working at low-temperature, comparatively simplicity in construction, high power density and ease of operation. The PEM fuel cell stack hardware makes use of Membrane Electrode Assembly (MEA), the bipolar plate, seal and end plate. For desirable currents and voltages, individual fuel cells are connected in series to form stacks of cells. Adjacent cells in a stack are typically separated by bipolar plates, which serve as the anode for one fuel cell and cathode for the next cell. Thus, the bipolar plate is a multi-functional component within a PEM fuel cell stack, that acts as a current collector, supply of reactant gases to the diffusion electrodes via flow channels and remover of water produced at cathode [1-2].

The effect of contaminants on fuel cells is one of the most important issues in fuel cells operation and applications [3]. It has been identified that out of fuel cell components the most affected by the contamination process is the membrane electrode assembly (MEA).Three major effects have been identified such as kinetic effect (poisoning of the electrode catalyst), conductivity effect (increase in

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the solid electrolyte resistance, including that of the membrane and catalyst layer ionomer and mass transfer effect (catalyst layer structure and hydrophobicity changes causing a mass transfer problem) [4]. However, various impurities in hydrogen fuel, such as CO, H_2S, NH_3, organic sulfur-carbon, and carbon-hydrogen compounds, and in air, such as NO_x, SO_x, and small organics, are brought alongwith the fuel and air feed streams into the anodes and cathodes in a PEM fuel cell stack, causing performance deterioration, and sometimes permanent damage to the MEAs [5, 6]. The hydrogen fuel impurities mentioned above mainly arise from the manufacturing process, in which natural gas (CH_4) or other organic fuels are reformed to produce hydrogen gas.

Park and O'Brien [5] and Hayter [6] briefly reviewed the effects of contaminations in the fuel and air streams on cell performance. They discussed CO, CO_2, NH_3, H_2S and inert diluents such as N_2 with respect to anode contaminations; SO_x and NO_x (cathodic contamination); and NH_3 and metallic species leached from the cell components (membrane contamination). Since then, a considerable amount of work has been carried out by many researchers who focused on the impacts of contaminations on fuel cell performance and lifetime.

It has been investigated extensively by many workers that hydrogen sulphide is an even more severe fuel contaminant than CO [7-10]. A trace amount of H_2S when exposed to an anode or cathode of the cell, was found to degrade the cell performance significantly mainly through the poisoning effect of the Platinum Catalysts [8]. Knights [4] found that at 100 mA cm^{-2} and 80 ^oC, 1.2 ppm H_2S could cause a cell voltage drop greater than 300 mV within 25 hour. This poisoning effect was also sensitive to the cell operation temperatures and load levels [4, 10]. Large performance losses were measured at H_2S concentrations as low as 50 ppm at 70 ^oC when the fuel cell anode was exposed to the H_2S-containing fuel for 3.8 hours. Unlike the case of CO poisoning, the presence of Ru in the platinum catalyst could not provide sufficient tolerance to H_2S poisoning [7].

Natural gas (the major part is methane) is one of the lightest hydrocarbon and promising candidate for hydrogen production. It is also available abundantly and being utilized as the preferred fuel in most of the countries. But the inherent problem with the natural gas is that it contains sulphur generally in the form of H_2S and some sulphur-containing tetrahydrothiophene (THT) for safety reasons. These compounds have poisoning effects for the hydrogen reforming catalysts and the catalysts deposited on the fuel cell electrodes. These also shorten the life of gas handling, storing and piping systems. Hence, it is necessary to remove these sulphur compounds from the raw natural gas or feedstock prior to reforming as well as from the product hydrogen gas due to multidimensional poisoning effects [11, 12].

Natural gas in pure form is an odourless gas and can not be detected in case of leakage. Tetrahydrothiophene (THT) is an induced impurity in the natural gas which is added in the pipeline before distribution to make it detectable by its bad smell. But for hydrogen fuel reforming the reaction takes place in the temperature range of 700 ^oC to 900 ^oC while the thermal decomposition of THT takes place into H_2S, thiophene and other sulfur compounds (S_o to S_{14}) in the temperature range of 450 ^oC to 500 ^oC [13].

The removal of sulfur from the natural gas is not an easy task and lot of efforts have been made with various techniques by many researchers and still the research work is in progress due to the limitations of every process. A number of sulfur-removing systems are and have been in use in different industrial as well as laboratory applications. As every process employed can remove one or more sulfur compounds and not all of them, the initial selection of a particular process may depend on feed parameters such as composition, pressure, temperature and nature of impurities as well as product specifications. The final selection of a particular process may be based on acid/sour gas percentage in the feed, whether only H_2S, or mixed [14]. Catalytic oxidation of H_2S is also one of the processes to convert it into elemental sulfur which then get adsorbed on the internal surface of the activated carbon, thus, leading to a sulfur load of 120% by weight [15-19].

Another way to remove sulfur compounds is to convert the organic sulfur species to H_2S over a hydrodesulphurization catalyst. The next step is

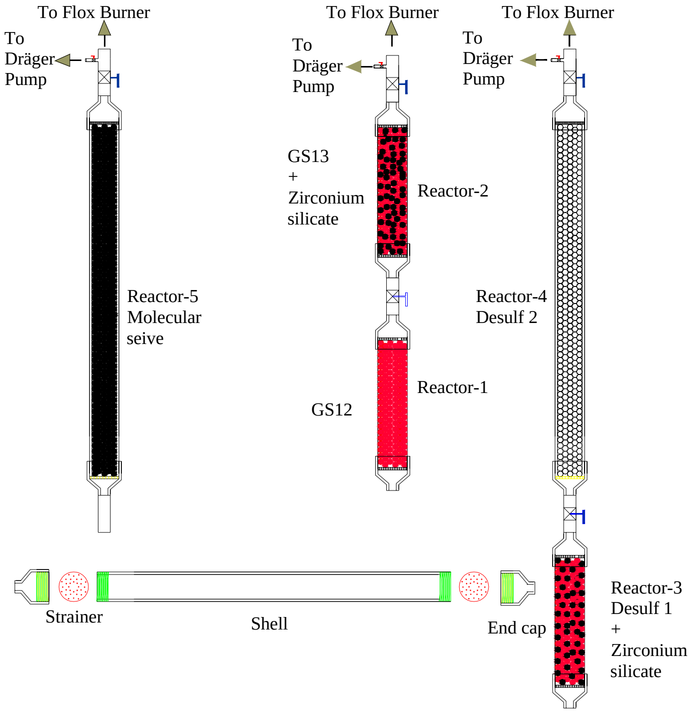

Figure 1. The reactors 1-5 alongwith assembly configuration and sorbent loadings.

sulfur removal with an absorbent. The same catalyst similarly converts any organo-chloride species to give HCl and also acts as an absorbent for most problematic metal species. A second absorbent is used for chloride removal [20-23]. For the removal of sulfur-containing odorants from natural gas for fuel cell-based combined heat and power applications, Beavon sulfur removal process, Co-capture of H_2S and CO_2 in pressurized gasifier-based process and calcium-based sorbents are employed [24]. The high sorption capacity of the sorbents has also been utilized for the removal of sulfur containing odorants from natural gas. Impregnated carbons containing several types of inorganic impregnants are also used for the adsorption of H_2S and mercaptans. Adsorption rates for H_2S as high as 50% by weight have been reported [25-27]. In one of the processes the gas containing H_2S is contacted with the solid sorbent to remove H_2S producing a product which has less than 0.1 ppm sulphur. This product is contacted with a highly selective reforming catalyst which is more sulphur sensitive under severe reforming conditions in subsequent reactors [28].

In the present communication, efforts were made to remove the sulfur compounds (H_2S and THT) from natural gas by the utilization of commercially available sulphur removing sorbents. The other aspect of the present investigation was to develop an economical and efficient desulphurization system for fuel cell applications. The main feature of this work was the replacement of costly sorbents by cheap molecular sieve, which shows an overall economical replacement. All results pertaining to the desulfurization of H_2S and THT are encouraging and are being reported in this paper.

2. Experimental

2.1 Materials

The reactor no. 1 and 2 (Fig. 1) were filled with the commercial sorbents made by company-A as follows; (1) The trade name of one of the sorbents filled in the reactor-1 was A1.1 with mass $M_{1.1}$ = 284 g and volume as 0.4 litre. (2) The 2nd sorbent filled in the reactor-2 was A1.2 alongwith the filler material (as it is first responsible for decomposition of the sulfur compounds and then the decomposed

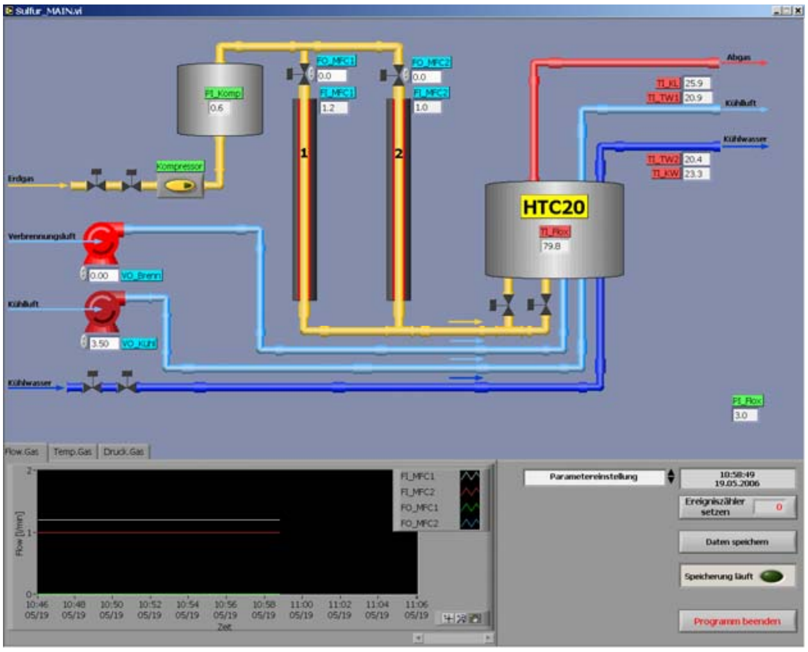

Figure 2. Experimental Setup for the removal of sulfur compounds.

components are absorbed by that material). Its mass was $M_{1.2}$ = 93 g and volume, 0.1 litre. The inert material zirconium silicate type Z volume, 0.3 litre. The reactor no. 3 was loaded with the sorbents made by company-B as follows; (3) The material $B_{2.1}$ was filled in the reactor no. 3 whose mass was $m_{2.1}$ = 159 g and volume, 0.17 litre. The volume of inert material (zirconium silicate) was 0.23 litre. The reactor no. 4 was filled with the sorbent material $B_{2.2.}$ whose mass was $m_{2.2}$ = 1037.9 g and volume was 1.11 litre. The inert material that is zirconium silicate (Sili Beads type Z) was used to fill and provide homogeneous space utilization.

The adsorbent, 13X molecular sieve ($Ca_{86}[(NaAlO_3)_{86}(SiO_2)_{106}]\cdot XH_2O$) was utilized in reactor no. 5 and its specifications are listed in Table 1.

2.2 *Instrumentation and analysis procedure*

The data equisition system (Fig. 2) was designed in the LAB View-6 program and installed on a P-4 computer. The computer was hooked up with the process controller which recieved inputs and out-puts from the pressure controllers, temperature controllers and flow controllers installed on the incoming lines of natural gas, combustion air, cooilng air and cooling water. To monitor and control the combustion of natural gas after cleaning, it was fed to the FLOX-burner whose exhaust was equiped with a gas analyzer. The parameters of the system were optimized and feed-back of the gas analyzer was utilized to control and monitor the flow of all of the supplies i.e., natural gas, air and water through sensors, solenoids and controllers. The respective parameters were adjusted to keep the combustion products e.g., CO_2, CO, NO_x and unburned CH_4 and O_2 within permisible limits as recomended by the Environmental Protection Agency, Germany.

Table 1. Specifications of 13X molecular sieve.

Properties	Values
Physical shape	Spheres
Size of spheres (mm)	Ø2.0-2.8
Wear ratio %≤	0.15
Bulk density g/ml	0.66
Ratio %	96
Static water adsorption %	24
Ethylene adsorbed %	18
Crushing strength (N/mm^2)	40
Coefficient of variation	0.3
Packing water %	1.5
Pore diameter (Å)	10
Regeneration temperature (^{o}C)	200-315

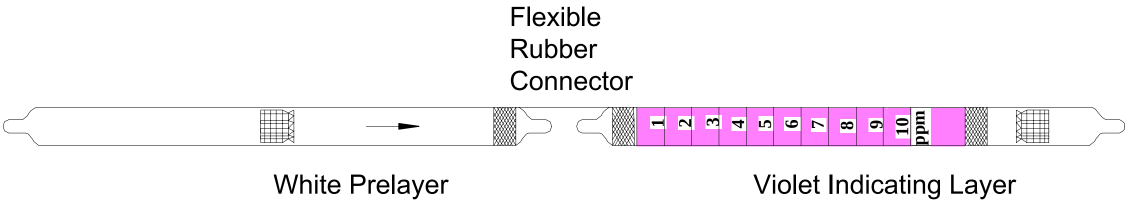

Figure 3. Dräger tubes for the determination and measurement of THT in the product gas.

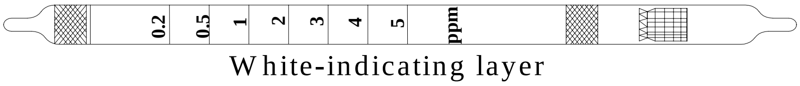

Figure 4. Dräger tube for the determination and measurement of H_2S in the product gas.

Figure 5. Dräger Accuro 2000 pump and tube system.

2.2.1 *Dräger sampling tubes*

Dräger sampling tubes (Figs. 3 and 4) and pump (Fig. 5) were used to perform identification and measurement of H_2S and THT present in the natural gas after the cleaning process. The contaminants in the natural gas were first collected by adsorption or chemisorption on silica gel. The sample was then analyzed both qualitatively and quantitatively by observing and comparing the change in colors of the adsorbed/chemisorbed species in different sections of the Dräger tube. The measuring range or detection limit of these tubes was 1-10 ppm (4–40 mg/m^3) for THT and 0.2-5 ppm for H_2S.

Following observation can be noted with the change of color as a result of reaction occurs;

$THT+KMnO_4\rightarrow$Yellowish brown reaction product

It sucks a calibrated 100 mL sample of gas through the Dräger-Tube® with each stroke. A built-in stroke counter displays stroke numbers have been made. The visual end-of-stroke indicator signifies the completion of each pump stroke.

2.3 *Development of desulphurization system*

The experimental desulphurization system (Fig. 2) consists of a compressor to store the natural gas in a small storage cylinder with a capacity of 25 litres and then this gas at a pressure of 1 bar and an average flow of 450 M^3/hour (at an ambient temperature of 25 ^oC) was supplied to two sets of sorbing reactors; each set having two reactors for different reactions.

The 5 reactors have the following dimensions; length of reactor no.1, no. 2 and no. 3 was 260 mm whereas 720 mm is the length of reactors no. 4 and 5. The outer diameter of all the reactors was

Table 2: Statistical data for the sorbents from company –A , Company – B and molecular sieve.

Run Time Hours	Reactors no. (1+2) (company –A)		Reactors no. (3+4) (company –B)		Reactor no. 5 13X Molecular Sieve	
	H_2S (ppm)	THT (ppm)	H_2S (ppm)	THT (ppm)	H_2S (ppm)	THT (ppm)
1	< MDL*	< MDL	< MDL	< MDL	< MDL	< MDL
50	< MDL	< MDL	< MDL	< MDL	< MDL	< MDL
100	< MDL	< MDL	< MDL	< MDL	< MDL	< MDL
576	< MDL	< MDL	< MDL	< MDL	< MDL	< MDL
580	< MDL	< MDL	< MDL	< MDL	< MDL	1
600	< MDL	< MDL	< MDL	< MDL	< MDL	10
946	< MDL	< MDL	< MDL	< MDL	< MDL	< MDL
950	< MDL	< MDL	< MDL	< MDL	< MDL	< MDL
1000	< MDL	8	< MDL	< MDL	< MDL	< MDL
1576	< MDL	< MDL	< MDL	< MDL	< MDL	< MDL
1580	< MDL	< MDL	< MDL	1	< MDL	< MDL
1600	< MDL	< MDL	< MDL	1.5	< MDL	< MDL
1816	< MDL	< MDL	< MDL	2	< MDL	< MDL
1864	< MDL	< MDL	< MDL	3	< MDL	< MDL
1900	< MDL	< MDL	< MDL	5	< MDL	< MDL
2000	< MDL	< MDL	< MDL	6	< MDL	< MDL

* < MDL (Less than the minimum detection limit, for H_2S= 0.2 ppm and THT= 1 ppm).

60 mm and inner diameter was 50 mm (Fig. 1). Each reactor was a tubular vessel of stainless steel with a thickness of 5 mm, having two strainers on each side (top and bottom) with holes of diameter 1.5 mm to keep the sorbents beads (Ø= 2-2.8 mm) within the reactor by allowing the natural gas to pass through the sorbent bed and two removable threaded end caps were provided on each side for sorbent loading and unloading.

The color coding of the piping system is indicated as follows:

Natural gas : Yellow (supplied through the compressed natural gas storage).

Air : Light blue (red color blower supply the combustion air and brown color blower supplies the cooling air).

Cooling water: Dark blue (water pump was used to draw the cooling water).

2.4. Procedure for determination of sulfur compounds

The reactors were operated by passing the pressurized natural gas into the reactor no. 2 through reactor no.1 and reactor no. 4 through reactor no. 3 in parallel (Fig.1) and after desulphurisation, the gas was supplied to a FLOX burner for safe disposal. In the meantime gas samples were taken from the outlet of the reactors no. 2 and 4 separately for making measurements and analysis of THT and H_2S present in the product stream at regular intervals of 4 hours.

At the outlets of the reactor no. 2 and 4 a nozzle of diameter 6 mm with gate valve (Fig. 1) was installed to get the sample of the natural gas for analysis after cleaning. The Dräger pump (model no: Accuro 2000) (Fig. 5) was used for sample suction from the desulphurised stream. The pump was pressed 5 times to make it clear of the previous readings. Then a Dräger pretube (Fig. 3) for H_2S adsorption was taken and its both ends were broken by inserting and twisting into the tube opener. The tube was installed into the nozzle on the outlet of the reactor no. 2 and 4 (Fig. 1) through the flexible rubber tube, one by one, by keeping the arrows towards the Dräger pump. This tube was joined with the THT determination graduated tube by directing the arrows towards the pump. Thirty strokes were applied through the Dräger tube as each pump stroke got 100 mL of sample gas. Thus, 3 litre of natural gas was

Table 3. Mass, volume and cost comparison of the 3 different materials applied.

Material Designation	Company–A		Cmpany–B		Molecular seive
Reactor No.	1	2	3	4	5
Material	A1.1	A1.2 + $ZrSiO_4$	B2.1+ $ZrSiO_4$	B2.2	13X
Mass (g)	284	93	159	1037.9	660
Volume (L)	0.4	0.1 + 0.3	0.17 + 0.23	1.11	1
Cost $ / L	14.18		48		1.75

passed through the Dräger tube. The Dräger tube was removed from the pump and after 10 minutes it was examined visually for any change in violet color of the graduated portion. However, careful observation confirmed that there was no change in color for the molecular sieve upto 580 hours run time in case of THT. Consequently THT was 1 ppm after 580 hours run time whereas the value of H_2S was less than the minimum detection limit of 1 ppm
(Table 2).

Similarly, the detection and measurement of H_2S in the natural gas were completed by using H_2S specific Dräger graduated tube (Fig. 4) after the completion of cleaning process. Both ends of this tube were opened by the help of tube opener and inserted into the pump, the tube was connected to the sample dispensing nozzle through the flexible rubber tube and 15 strokes were applied to get 1.5 litre of sample gas. After 15 minutes, the tube was carefully checked for any change in color and the presence of H_2S in the sample.

In this way measurement was taken every 4 hours run-time of the system for 2000 hours to check natural gas cleaning process and to determine the affective life cycle of the sorbent materials alongwith degradation behavior. The results were tabulated in Table 2 to evaluate the performance and life of the sorbents from company-A, company-B and that of molecular sieve.

3. Results and Discussion

A well-defined and economical way has been adopted to remove sulfur compounds from the feedstock natural gas for the production of clean gas which will meet the requirement of the fuel reforming systems and later on serves as a feed gas for fuel cell systems. The cost of the materials used initially in the reactor no.1, 2 (from company-A) and 3, 4 (company-B) were approx. Euro 25 per litre. Moreover the cost of the materials loaded in any pair of reactors along with relevant parameters is presented in Table 3 for comparison. Similarly the 13X molecular sieve costs at the rate of Euro 1.75 per litre (only one litre was loaded into the reactor no. 5) showing a significant running cost savings for the same flow of natural gas, same composition of gas and same H_2S and THT loadings (Table 4).

A desulphurization test system has been developed (Fig. 2) to ascertain the efficiency of the desulphurization sorbents and the design parameters of the reactors, etc. The natural gas (with various constituents and sulfur impurities) was analyzed by using the portable gas analyzer model Lancom III, AMETEK Inc. USA and resulting percentages of components are given in Table 4.

Table 4. Composition of natural gas supplied at ZSW lab. Germany.

Component	Volume %
Methane	97.88
Ethane	0.79
Propane	0.24
Butane	0.06
Pentane	0.01
Carbondioxide (CO_2)	0.012
Oxygen	0.9
Hydrogen sulphide (H_2S) (mg/m^3)	<1
Tetrahydrothiophene (THT) (mg/m^3)	18.1
Sulfur (in the form of H_2S and THT) (mg/m^3)	6.5

The amount of H_2S and THT was measured in the product gas stream with the help of Dräger Accuro 2000 pump and tube system as shown in

Fig. 5. The values of both these sulfur compounds were found less than the respective detection limits upto 576 hours run-time (Table 2), for molecular sieve loaded in the reactor no. 5 (Fig. 1). Even after 580 hours H_2S was found less than the minimum detection limit of 0.2 ppm but THT measured as 1 ppm rose to 10 ppm after the completion of 600 hours run-time. This showed that the molecular sieve got saturated and lost the efficiency for the sorption of THT, so monitoring was discontinued for regeneration process. It was noticed that the desulfurization cycle of 13X molecular sieve without regeneration was 500 hours thereby cleaning 225,000 M^3 of natural gas by applying the above quoted conditions.

It was observed that the channel diameters of 13X molecular sieve cages (2-9 $\underline{\text{Å}}$) were effective enough to adsorb H_2S and THT with reasonable capacity which could be utilized for the desulfurization of natural gas for fuel reforming processes that would lead to ultimate utilization of hydrogen in the fuel cells.

Similarly, the sorbents loaded into the reactors no. 1 and 2 (A1.1 and B1.2) performed well up to 946 hours run-time (Table 2) for H_2S and THT compounds and after 950 hours run time the H_2S was still less than the minimum detection limit of 0.2 ppm but THT was found 1 ppm that gradually increased to 8 ppm after the completion of 1000 hours. This showed that the sorption capacity of these materials was degrading continuously for THT on the completion of 950 hours run-time, so the measurements for these materials were stopped. It was noted that these materials were suitable for desulphurisation of 405,000 M^3 of natural gas as per operating conditions mentioned above.

On the other hand, the sorbents loaded into the reactors no. 3 and 4 (B2.1 and B2.2) were showing the concentration of H_2S and THT in the product gas stream less than the minimum detection limits upto the run time of 1576 hours. After 1580 hours run-time (Table 2) the H_2S was still less than the minimum detection limit of 0.2 ppm but THT was found 1 ppm and gradually it soared to 6 ppm after the completion of 2000 hours. Hence further measurements were discontinued. It was clear that B2.1 and B2.2 are satisfactory for 1500 hours run-time cleaning 675,000 M^3 of natural gas with above-mentioned operating conditions.

In the same way it was proved that with the operating conditions maintained B2.1 and B2.2 have the desulfurization capacity of 675,000 M^3 without regeneration costing 48 Euro and the volume of natural gas cleaned per Euro earned out to be 14000 M^3. A1.1 and A1.2 have the desulfurization capacity of 405,000 M^3 before regeneration and these cost 14.18 Euro, giving volume of natural gas cleaned per Euro as 28561 M^3. Finally the 13X molecular sieve has the desulfurization capacity of 225,000 M^3 before regeneration and it costs 1.75 Euro, the volume of natural gas cleaned per Euro was 128571 M^3 which is maximum as compared to the other two sets of sorbent materials (Table 3).

4. Conclusion

The efficiency of the three materials are comparable and within acceptable limits but the construction design of desulphurization plant from company-A and company-B is comparatively complex as compared to the system required by 13X molecular sieve. Moreover, the running cost of molecular sieve is reasonably less than that of the other two materials. Hence, 13X molecular sieve is recommended as economical replacement.

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References

[1] H. Tsuchiya and O. Kobayashi, Int. J. Hydrogen Energy, **29** (2004) 985.

[2] D.P. Davies, P.L. Adcock, M. Turpin and S.J. Rowen, J. Power Sources., **86** (2000) 237.

[3] J. Zhang, H. Wang, D. P. Wilkinson, D. Song, J. Shen and Z.S. Liu. J. Power Sources, **147** (2005) 58.

[4] S. Knights, N. Jia, C. Chuy and J. Zhang, Fuel Cell Seminar 2005: Fuel Cell Progress, Challenges and Markets, Palm Springs, California, 2005.

[5] S. M. Park and T. J.O. Brien, Technical Report (# DOE/METC/RI-80/16), Department of Energy, Morgantown, WV, USA, 1980.

[6] P.R. Hayter, P. Mitchell, R.A.J. Dams, C. Dudfield and N. Gladding, Contract Report (ETSUF/02/00126/REP), Welman CJB Limited , Portsmouth, UK,1997.

[7] M. Murthy, M. Esayian, W. K. Lee and J. W. Van Zee, J. Electrochemical. Soc. **150** (2003) A29.

[8] R. Mohtadi, W. K. Lee, S. Cowan, J. W. Van Zee and M. Murthy, Electrochem, Solid-State Lett. **6** (2003) A272.

[9] M.J. Escudero, S. Jimenez and L. Daza, Proceedings of the First European Fuel Cell Technology and Applications Conference 2005, Book of Abstacts (2005) P. 169.

[10] R. Mohtadi, W.K. Lee, S. Cowan and J.W. Van Zee, Appl. Catal. B: Environ. **56** (2005) 37.

[11] X. Cheng, Z, Shi, N. Glass, L. Zhang, J. Zhang, D. Song, Z.S. Liu, H. Wang and J. Shen, J. Power Sources, **165** (2007) 739.

[12] W. Shi, B. Yi, M. Hou, F. Jing, H. Yu and P. Ming, J. Power Sources **164** (2007) 272.

[13] D. Xia, Y. Tian, G. Zhu, Y. Xiang, L. Luo, and Thomas T.-S. Huang, J. Energy & Fuels, **21** (2007) 1.

[14] M. Rameshni, Parsons E&C Manual, (2000) 2.

[15] Aminex, Hydrocarbon Processing- Gas Processing Handbook, (1984) 87.

[16] Mericat II, Hydrocarbon Technology International (1993).

[17] J. Klein and K.D. Henning, Fuel **63** (1984) 1064.

[18] Le Strat, P. Y., Oil and gas Journal (2001) 46.

[19] T. Regen, Oil and Gas Journal, (1985) 78.

[20] Method for Removal of Acid Gases from a Gas Flow, U.S Patent No.7004997.

[21] M.S. Horikawa, F. Rossi, M.L. Gimenes, C.M. M. Costa and M.G.C. da Siva, J. Chem. Eng. **21**, No. 3 (2004) 1.

[22] Hydrogen feedstock purification, Johnson Matthey Catalysts, Technical Information, http://www.jmcatalysts.com/pct/marketshome.asp?marketid=15& id= 288

[23] J.M. Bremmer and W.L. Banwart, Soil Bio. and Biochem., **8** (1976) 79.

[24] D. M. Fenton and H. W. Gowdy, J. Environ. Inter., **2** (1979) 183.

[25] P. Sun, John R. Grace, C. Jim Lin and E.J. Anthony, J. Energy and Fuels **10** (2007) 510.

[26] G.O. Alptekin, Sixth Annual SECA Workshop, Pacific Grove, California, April 21, 2004.

[27] Ho-Jung Ryu, John R. Grace and C. Jim Lim, Energy and Fuels, **20** (2006) 1621.

[28] H. T. Kim, K. W. Jun, S. M. Kim, H. S. Potdar and Y. S. Yoon, Energy and Fuels (2006) 236.