Proceedings of The Canadian Society for Mechanical Engineering International Congress 2018
CSME International Congress 2018
May 27-30, 2018, Toronto, On, Canada

Adsorbent-Adsorbate Pairs for Solar Thermal Energy Storage in Residential Heating Applications: A Comparative Study

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Abstract— This paper investigates the feasibility of using different adsorbent-adsorbate pairs in a thermal energy storage cycle to store solar energy for residential heating applications in Canada. Silica gel, activated carbon, activated aluminum, zeolite-4A, zeolite-5A and zeolite-13X adsorbents paired with methanol and water adsorbates are considered. Calculations are made to determine the volume, mass and cost of the adsorbent-adsorbate pair required to heat a house with four occupants. Zeolite 4A-water and zeolite 13X-water pairs are found to be the most economic (with an actual cost of 285 CAD and 374 CAD, respectively) and efficient (maximum heat of adsorption) adsorbent-adsorbate pairs with the minimum mass required, (290 kg and 226 kg, respectively) to meet the spatial heating requirements of the house.

Keywords- Adsorption, adsorption pairs, thermal energy storage

I. INTRODUCTION

Policy changes incentivizing sustainable energy technologies have provided new opportunities for renewable sources of energy such as solar, wind, tidal and others. Investments in solar energy, which is abundantly available, has increased exponentially in recent years. However, solar energy is intermittent and should be coupled with energy storage technologies to reach its full potential. For example, one promising technology for utilizing solar energy in a Canadian climate is to store it in a thermal battery so it can be used at night to reduce building heating loads. As solar thermal energy storage costs are considerable, various technologies are being explored to optimize its capacity, operating temperature, and overall performance. Heat energy is stored at different temperatures depending on its intended application. Low temperature heat is useful for domestic air-conditioning applications whereas high temperature heat is extremely useful in industry. Furthermore, heat energy can be stored in various forms: sensible heat (solids, liquids, solid-liquid), latent heat (solid-solid, solid-liquid), chemical heat (reacting solids or liquids, gaseous compounds) and the heat of adsorption or absorption (physisorption, chemisorption, heat of solutions). Sensible heat storage technologies utilize high heat capacity materials, whereas latent heat energy storage technologies absorb/release heat when a material undergoes a phase change from solid to liquid or liquid to gas and vice-versa. On the other hand, chemical heat energy storage technologies utilize reversible thermo-chemical reactions (Rainer et al. 2012) to absorb and release heat. Sorption-based thermal storage technologies can be broadly categorized as absorption and adsorption processes, depending on whether the adsorbate atoms or molecules are dissolved in, or adhered to, the surface of the absorbent. Advantages of sorption-based thermal energy storage technologies are their minimal costs, and matured performance in heat pumping and solar refrigeration. In this article, adsorption-based thermal energy storage materials are investigated for their potential to store solar energy during the day to provide heat at night in a typical Canadian residential home.

II. LITERATURE REVIEW

Studies have been conducted to understand thermal energy storage technologies such as electric thermal storage heaters, salt hydrate technology, molten salt technologies and solar energy storage. (Bataineh and Taamneh 2016) conducted a feasibility analysis of both adsorption and absorption systems and concluded that extended adsorption and desorption times, poor exhibitions, and low coefficient of performance (COP) are hindering the widespread commercialization of these technologies. (Papadopoulos et al. 2003) reviewed existing solar energy based sorptive refrigeration technologies for applications in both residential and commercial spaces, and discussed the necessity of implementing energy policies to accelerate the flourishment of solar refrigeration technologies. (Cabeza et al. 2017) presented an exhaustive review on adsorption and absorption technologies in refrigeration and thermal heat storage and discussed research progression towards developing efficient working pairs with better storage capacities and optimal energy output. (Aliane et al. 2016) studied various solar energy technologies developed for adsorptive cooling. A comparative study of various experimental systems has been done to study the operative behaviors of individual components and whole systems. It was concluded that these technologies can potentially be extended to meet global energy demands by implementing some modest improvements. Further, (Alobaid et al. 2017) reviewed solar absorption cooling systems which were fifty percent more energy efficient than vapor compression based cooling systems. The COP and thermal collector efficiency of the solar based systems are found to vary from 0.1-0.91 and 0.06-0.64, respectively. It was also noted that solar collector costs comprise a significant portion of the entire system. (Zeyghami et al. 2015) discussed various concepts, designs, and experimental set-ups for thermomechanical solar adsorption systems while discussing performance enhancement approaches. (Goyal et al. 2016) presented the fundamentals of solar energy utilization in adsorption systems and provided a thorough discussion about technological and economic aspects, recognizing that poor heat and mass transfer performances, low COP, and higher costs are the major roadblocks preventing the development of competitive products in this technological field. It has also been observed that hybrid systems, which combine solar adsorption heating and cooling, enhance the system performances significantly. (Askalany et al. 2013) compared various adsorption cooling systems and presented a comparision based on adsorbent-refrigrent pairs on the basis of COP, specific cooling power, driving and evaporation temperatures. It has been observed that the systems employing silica gel-water and chloride composites-water pairs have the highest COP, while those employing zeolite-water pairs have the lowest COP. The lowest evaporation temperatures were found in a system that used metal hydride-water pairs and the lowest driving temperatures were observed for the case of silica-gel-methanol and chloride composites-methanol pairs, wheras the highest driving temperatures are found in zeolite-water pairs.

III. ADSORPTION

Adsorption involves the adhesion of adsorbate molecules on the adsorbent surface. The adsorbent is usually a porous material with a high surface area. Adsorbate molecules form a thin film on the surface of the adsorbent either by physisorption or chemisorption. Adsorption is referred to as physisorption if the adsorbate molecules are attached on the adsorbent surface by Vander Waals forces, and chemisorption if the adsorbate molecules are chemically attached at the adsorbent surface. Thermal heat storage, therefore, is a physio-chemical phenomenon between the adsorbate and adsorbent molecules while condensation and evaporation facilitate the charging and release of adsorbate molecule onto and from the adsorbent, respectively.

IV. THERMAL ENERGY STORAGE CYCLE

The thermal energy storage cycle for the application of residential heating in Canada, shown in Figure 1, comprises two main processes: desorption and adsorption.

***Desorption process*:** During the day the solar irradiance provides the heat of adsorption required to drive the evaporation of adsorbate molecules from the adsorber. The average daily solar insolation available from November through March in Toronto, Canada is 7.8 MJ/m^2 (October-9.3 MJ/m^2, November-5.1 MJ/m^2, December-4.3 MJ/m^2, January-5.7 MJ/m^2, February-9.2 MJ/m^2 and March-13.2 MJ/m^2) [26]. Assuming the average size of a residential building with four occupants is 200 m^2 [27] the net solar radiance available during these cold climatic conditions is 1.56 GJ per day.

Adsorption process: During the night adsorbate molecules that have evaporated from the condenser/evaporator are adsorbed by the adsorbent, and the heat of vaporization is released to function as a heat supply for residential heating. The reaction that occurs during the desorption and adsorption processes can be written as follows:

$$Adsorbent + n{\cdot}Adsorbate \leftrightarrow Adsorbent * n{\cdot}Adsorbate + Heat$$

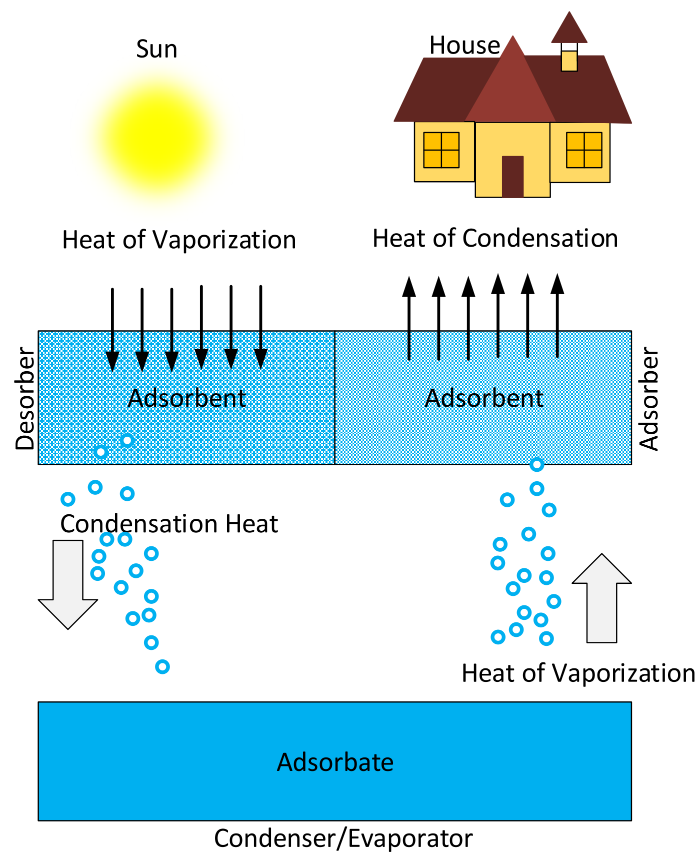

Fig 1. Schematic diagram showing the desorption (left) and adsorption (right) processes in the thermal energy adsorption cycle.

V. ADSORPTION PAIRS

The adsorbate and adsorbent form an essential component of the thermal energy storage cycle and various factors play a decisive role in selecting the right pair including the maximum adsorption capacity, heat of adsorption, energy capacity, specific heat of adsorbent, adsorbent density, critical pressure and critical temperature. The maximum adsorption capacity is the maximum amount of adsorbate an adsorbent can adsorb. The heat of adsorption is the heat released during the adsorption process, and can be determined by estimating the heat released in a calorimeter experiment or by adsorption isotherms. The energy capacity is the maximum energy an adsorbent can absorb or release. The specific heat of adsorbent is the heat required to raise the temperature of the adsorbent by one degree under constant pressure. At the critical temperature and critical pressure, the density of the adsorbate liquid and vapor is equal.

While many different adsorbates are available for various applications, the most commonly used are water, ammonia, methanol and ethanol. The latent heat of evaporation of water (2258 kJ/kg) is very high compared to that of ammonia (1368 kJ/kg), methanol (1100 kJ/kg) and ethanol (1100 kJ/kg). Thus, water is typically preferred, although other adsorbates can be used to produce sub-zero temperatures. Activated carbons, zeolites, silica gels, metal-organic frameworks, and potassium hydroxide (KOH) are widely used adsorbents as they have a highly porous structure, which increases the adsorption capacity of the system. (Aristov 2007) presented selective water sorbents based on halides, sulphates and nitrates of alkaline and alkaline earth metals possessing an intermediate behavior between solid adsorbents, salt hydrates, and liquid absorbents. Also, an estimation was made on the thermodynamic equilibrium of these materials with water vapors. These sorbents are found to possess a better COP then silica gel-water and zeolite-water based systems. (Aristov 2013) reviewed existing literature on metalaluminophosphates, metal-organic frameworks, ordered porous solids, and porous carbons and composite frameworks. (Gordeeva and Aristov 2012) surveyed composite salts inside porous matrix (CSPM) sorbents. (Bhargav et al. 2017) studied the methanol-activated carbon fiber adsorption pair, and the adsorption capacity and desorption temperature were investigated under isobaric conditions using the Dubinin–Astakhov equation. (Brancato et al. 2015) tested various activated carbon fibers and composites of LiBr on silica gel adsorbents along with ethanol as a refrigerant. A thermo-physical analysis including nitrogen physisorption, specific heat and thermo-gravimetric equilibrium has been performed, and the Dubinin–Astakhov equation has been used to fit the equilibrium data. Furthermore, the thermodynamic performance was estimated by calculating the COP. (Erto et al. 2010) studied trichloroethylene adsorbates precipitated from water-based solutions on a set of 12 different activated carbon adsorbents. Results indicated that the adsorption capacity increases with an increase in Brunauer-Emmett-Teller (BET) surface area, micropore volume, and the percentage of carbon in the adsorbent. Further, the presence of sodium and tetrachloroethylene also increased the absorption capacity. (Meng and Park 2010) improved the CO_2 adsorption capacity by inserting KOH in graphite nanofibers to increase its porosity under heat treatment at 700-1000℃. The heat treatment temperature had a significant effect on the adsorption capacity and texture of graphite nanofibers, and a positive effect on surface area, total pore volume and micropore volume. (Saha et al. 2011) studied the adsorption of an ethanol-MIL101Cr pair theoretically and experimentally within a 0.1-0.9 bar pressure range and a 30-70°C temperature range. One kg of MIL-101 Cr is found to adsorb up to 1.1 kg of ethanol at 30°C. The Tóth equation has been used to fit experimental data with the goal of enhancing the adsorption capacity in the adsorption cooling cycle employing activated carbon fibers (ACF) as adsorbents. (Saha et al. 2015) studied various adsorbent-refrigerant pairs at a temperature of 77.3K. ACF (A-20), owing to its large surface area, MIL-101Cr was found to have 0.797 kg/kg ethanol adsorption capacity. (Uddin et al. 2014) studied the highly porous Maxsorb III, H_2 and KOH-H_2 surface treated Maxsorb III adsorbents for ethanol adsorption at 30-70°C and evaporation temperatures of 65°C experimentally. The Dubinin-Radushkevich and Dubinin-Astakhov adsorption isotherm models have been used to obtain adsorption isotherms. Adsorption cycle performance studies were conducted using activated carbon-ethanol adsorption pairs. H_2 surface treated Maxsorb III exhibits a COP of 0.51 with a specific cooling effect of 374 kJ/kg at evaporation, heat source, and heat sink temperatures of -5°C, 100°C and 30°C, respectively. (Zeng et al. 2017) studied the ammonia adsorption on four different kinds of activated carbons at 30°C. MSC30 is found to possess a very high adsorption capacity for ammonia. A modified Dubinin-Astakhov equation has been used for isothermal studies. (Dawoud et al. 2007) experimentally studied an adsorption system using a zeolite 13X-water adsorption pair. It was observed that an increase in the flow rates of the adsorber and desorber increase the heat discharge, while higher temperatures occured at lower flow rates. A radiation shield is found to be effective in minimizing radiation losses during the adsorption process. (Ansari et al. 2014) prepared nano-NaX zeolite using a hydrothermal process. The size and crystallinity of the NaX zeolites is found to increase up to 44 nm and 96%, respectively, as the reaction time was increased from 1h to 4h and the reaction temperature increased from 90°C to 110°C. (Tatler and Erdem-Şenatalar 2004) evaluated the effective diffusion coefficient of water in zeolite-4A coatings. It is observed that the evaluated results qualify the thermogravimetric analysis (TGA) and effective medium theory (EMT) based experimental results from the literature. (Solmuş et al. 2011) developed a zeolite-water pair based adsorption cooling system and investigated its performance at various evaporator temperatures. The COP of the system was about 0.25 at adsorption, desorption, and condenser temperatures of 45°C, 150°C, and 30°C, respectively, for evaporative temperatures ranging from 10°C to 22.5°C. The mean volumetric specific cooling power density and the mean mass specific cooling power density were found to be 4.8 kW/m^3 and 6.4 W/kg, respectively. (Solmuş et al. 2010) used the Dubinin-Astakhov equation to determine a maximum adsorption capacity of 0.12 kg_w/kg_{ad} for a natural zeolite. Furthermore, the isosteric heat was calculated and a correlation between adsorption and desorption processes was established.

VI. DISCUSSION

Herein, different adsorbate-adsorbent pairs are investigated for their ability to store solar energy during the day to provide heating for a typical residential home in Canada. The average size of a residence with four occupants is around 200 m^2 [26] with the overall energy requirements of 130 GJ yearly, or 0.36 GJ daily, by Statistics Canada [27], and it is assumed that 63% of this energy is used for spatial heating [28]. Thus, it is assumed that 0.23 GJ is required for spatial heating, which is much less than the average solar irradiance (1.6 GJ) estimated to be incident on a residential home in Toronto over the months from November to March (Table III).

Different adsorbate-adsorbent pairs comprising seven adsorbents (charcoal, silica gel, activated alumina, zeolie-4A, -5A, -13X, and activated carbon) with water and methanol as properties of adsorbate-adsorbent pairs are listed in Table I. The adsorbate capacity (figure 3) for the adsorbents considered herein varies from 0.16-0.37 $kg_{adsorbate}/kg_{adsorbent}$, `with the silica gel-water having the maximum followed by active-carbon at 0.32 $kg_{adsorbate}/kg_{adsorbent}$ and zeolite-13X at 0.3 $kg_{adsorbate}/kg_{adsorbent}$ whereas the heat of adsorption (figure 4) of zeolite is the largest with a value of 1400-4410 kJ/$kg_{adsorbate}$. Here the zeolite 4A-water and zeolite 13X-water pairs show high potential with energy capacities of 970 and 1320 kJ/$kg_{adsorbent}$, respectively, which is the largest amongst all pairs considered. Thermodynamic properties viz. molecular weight, critical temperature, critical pressure, density and specific heat of some selective adsorbates for adsorption based cooling and heating are given in Table II. Further, Table I shows the net heat of adsorption (figure 8) per mass of adsorbent is the largest for zeolite 4A-water and zeolite 13X-

TABLE I. THERMOCHEMICAL PROPERTIES OF ADSORBENT–ADSORBATE PAIRS (RAINER ET AL. 2012)

Material	Adsorbate	Max. adsorbate capacity	Heat of adsorption (average)	Adsorbent specific heat	Net heat of adsorption	Net Heat of adsorption per unit volume of adsorbent	Volume of Adsorbent required for residential heating	Net adsorbent required	Amount of Adsorbate required	Total mass of system
			Δh_{ads}	$c_{adsorbent}$						
		$kg_{adsorbate}/kg_{adsorbent}$	kJ/$kg_{adsorbate}$	kJ kg^{-1} K^{-1}	kJ/$kg_{adsorbent}$	kJ/m^3	m^3	kg	kg	kg
Charcoal	**Water**	0.4	2320	1.09	928	445440	0.52	247.84	99.14	346.98
Silica gel	**Water**	0.37	2560	0.88	947.2	634624	0.36	242.82	89.84	332.66
Activated alumina	**Water**	0.19	2480	1	471.2	461776	0.50	488.12	92.74	580.86
Zeolites 4A	**Water**	0.22	4410	1.05	970.2	756756	0.30	237.06	52.15	289.22
Zeolites 5A	**Water**	0.22	4180	1.05	919.6	625328	0.37	250.11	55.02	305.13
Zeolites 13 X	**Water**	0.3	4410	0.92	1323	793800	0.29	173.85	52.15	226.00
Active carbon	**Methanol**	0.32	1400	0.9	448	-	-	-	-	-
Zeolites 4A	**Methanol**	0.16	2300	1.05	368	287040	0.80	625.00	100.00	725.00
Zeolites 5A	**Methanol**	0.17	2300	1.05	391	265880	0.87	588.24	100.00	688.24
Zeolites 13 X	**Methanol**	0.2	2400	0.92	480	288000	0.80	479.17	95.83	575.00

TABLE II. THERMODYNAMIC PROPERTIES OF ADSORBATES FOR ADSORPTION COOLING AND HEATING [17]

	Mol wt.	Critical temp.	Critical pressure	Density	Specific heat
		T_{cr}	P_{cr}	ρ_{cr}	$C_{p0,cr}$
	g mol^{-1}	K	kPa	mol m^{-3}	kJ mol^{-1} K^{-1}
Methanol	32.042	512.64	8140	8547	0.061
Ethanol	46.069	513.92	6120	5952	0.098
Ammonia	17.031	405.65	11300	13889	0.038
Water	18.015	647.3	22048	17857	0.037
1-Propanol	60.096	536.78	5120	4545	0.135
2-Propanol	60.096	508.3	4790	4525	0.133

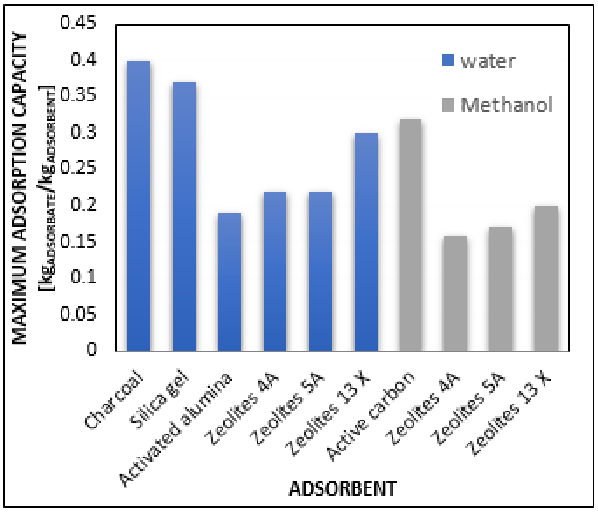

Fig 2. Maximum adsorption capacity of various adsorbents

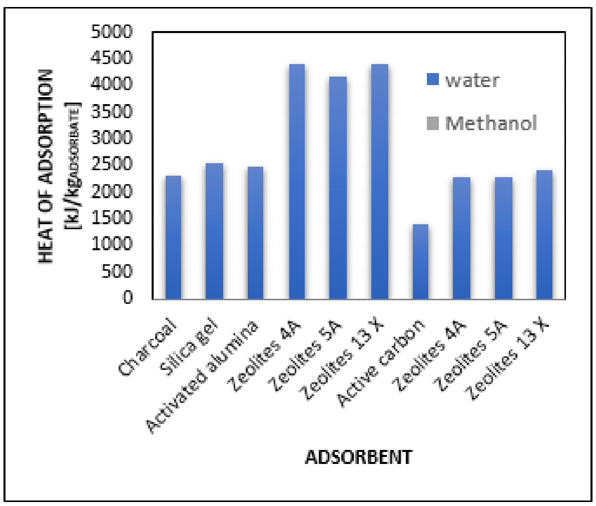

Fig 3. Heat of adsorption generated with one kilogram of various adsorbents

TABLE III. AVERAGE ENERGY REQUIREMENTS OF A HOUSE OF FOUR IN CANADA [SOURCE: STATISTICS CANADA]

For a house of 4 occupants with an area 206.4 m²	GJ
Average energy requirement yearly	130
Average energy requirement per Day	0.36
Average energy requirement for spatial heating per Day	0.23
Average solar energy available per day in Toronto (November-March)	1.56

water with values of 970 and 1323 kJ/$kg_{adsorbent}$, respectively. The total volume of adsorbent required to provide heating for a residential building with four occupants in Canada using a zeolite 4A-water pair is 0.30 m^3, while the volume would be 0.29 m^3 if a zeolite 13X-water pair were to be used. If a zeolite

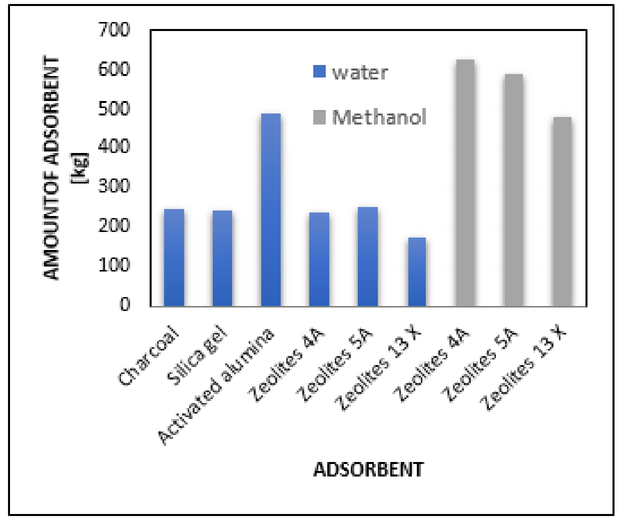

Fig 4. Amount of adsorbents needed to meet the energy requirements for a house with four occupants in Canada

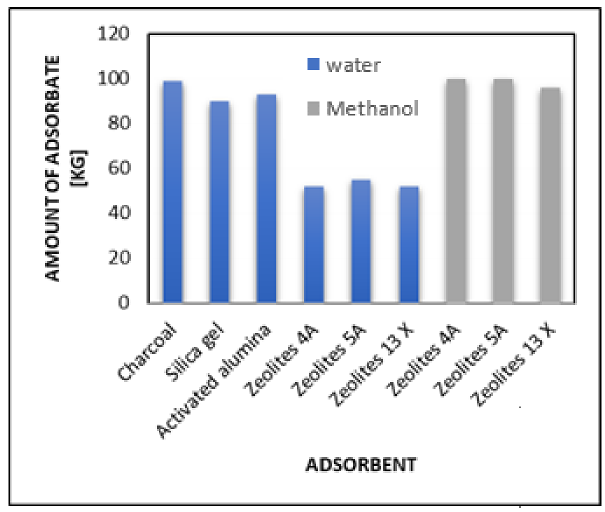

Fig 5. Amount of adsorbate needed to meet the energy requirements for a house with four occupants in Canada

5A-methanol pair were used as the adsorbent/adsorbate then the required volume would increase significantly to 0.87 m^3. Also, zeolite 4A-water and zeolite 13X-water pairs have the minimum mass of adsorbent (figure 5) and adsorbate (figure 6) required. The total mass of the adsorption system including both adsorbent and adsorbate needed to meet the energy requirement of the house under consideration for zeolite 4A-water and zeolite 13X-water is 290 kg and 226 kg, respectively. Now, if the adsorbent-absorbent pair is situated inside flat rectangular panels that are 1.7 m long, 1 m wide and 3 cm thick, it would take only 6 panels of zeolite 4A-water and 6 panels of zeolite 13X-water pairs to meet the spatial heating requirements. The total cost of the adsorbents in discussion is shown in Figure 9 and the cost of zeolite 4A and zeolite 13X adsorbents required to meet the energy for the house under consideration is 285 CAD and 375 CAD,

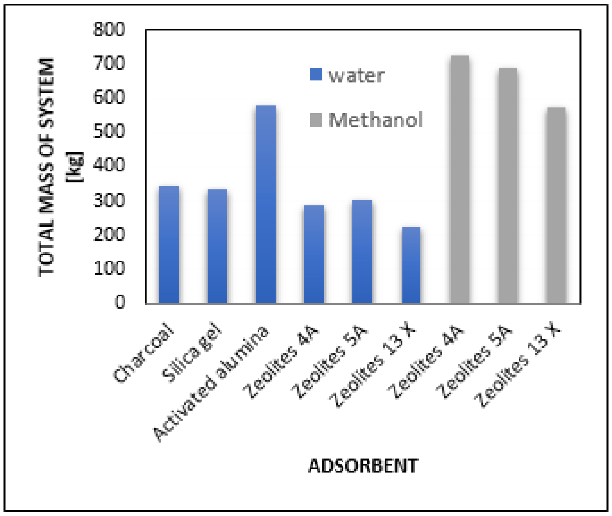

Fig 6. Total mass of the system (adsorbent and adsorbate) needed to meet the energy requirements for a house with four occupants in Canada

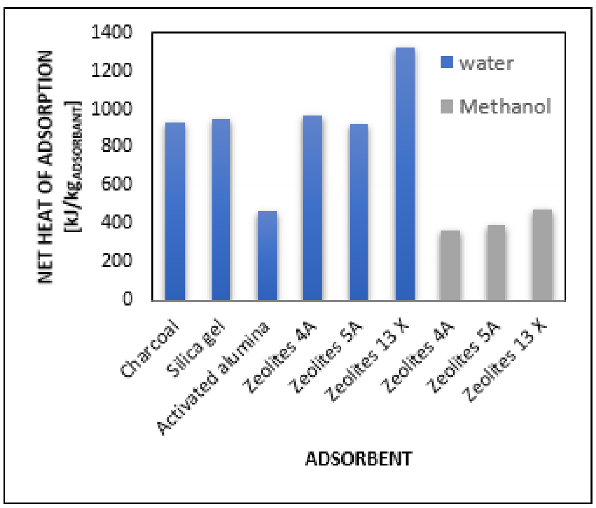

Fig 7. Net Heat of adsorption of various adsorbents

In comparison, an average of about 6.2 m^3 [29] of natural gas per day is required to meet the spatial heating requirements for the residence under consideration, and the daily cost of using natural gas for spatial heating would be about 0.6 CAD. Further, assuming a carbon tax of 1 CAD per GJ [31] of energy produced, this cost would rise from 0.6 CAD to 0.85 CAD daily (~19 CAD monthly). Thus, the owner of the residence under consideration would be able to save ~155 CAD on their natural gas heating bill over the colder seasons of the year by using adsorptive technology to heat their home. Perhaps more importantly, considering CO_2 emissions, 1 GJ of energy generation from natural gas emits approximately 56 kg of CO_2 [30]. For the residence with four occupants and a daily energy requirement of 0.23 GJ, ~13 kg of CO_2 is emitted daily to provide for heating. Thus, about 2.4 tonnes of CO_2 emissions could potentially be prevented by heating a typical

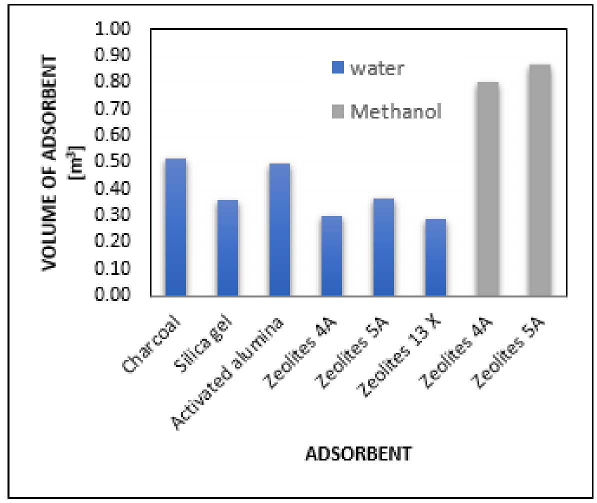

Fig 8. Volume of adsorbents needed to meet the energy requirements for a house with four occupants in Canada

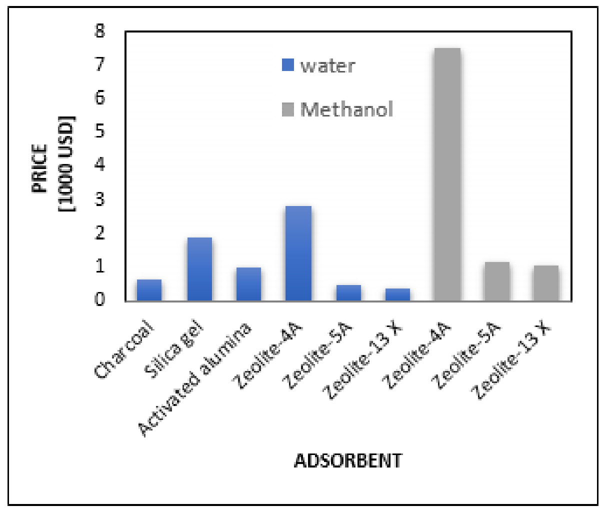

Fig 9. Costs of various adsorbents

Canadian residence using a thermal energy storage cycle based on adsorbate-adsorbent pairs instead of using natural gas. As CO_2 emissions per capita in Ontario are ~15 tonnes, further research and development of sorption-based thermal energy storage cycles that store solar energy for residential heating applications is warranted.

VII. CONCLUSION

Various adsorbent-adsorbate pairs have been investigated as the working pair in a thermal energy adsorption cycle that provides the energy required to heat a residence with four occupants in Canada. Amongst all pairs considered, the zeolite 4A-water and zeolite 13X-water pairs have the maximum heat of adsorption and the minimum mass (290 kg and 226 kg, respectively) required to meet the heating demands. The net volume of zeolite 4A and zeolite 13X required to meet the spatial heating requirements of the residence is 0.29 m^2 and

0.30 m^2, respectively. The cost of zeolite 4A and zeolite 13X adsorbents for meeting the heating energy requirements of the residence is 285 CAD and 374 CAD, respectively. Thus, zeolite 4A-water and zeolite 13X-water pairs can potentially be used in a thermal energy adsorption cycle that stores solar energy so it can be used to provide heating for a residential home in Canada and further research in this area is warranted.

VIII. ACKNOWLEDGEMENTS

This research was supported by the Natural Sciences and Engineering Research Council of Canada

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