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Comparative Thermodynamic Study of Five Couples Used in Solar Cooling with Adsorption by Simulation

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Authors' contributions

This work was carried out in collaboration between all authors. Authors AK and DJB designed the study, performed the statistical analysis, wrote the protocol, and wrote the first draft of the manuscript and managed literature searches. Authors SK and MO managed the analyses of the study and literature searches. All authors read and approved the final manuscript.

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ABSTRACT

Aims: The aim of this work was to develop a thermodynamic model that applies to several couples, for determining the $_{COP_{ik}}$ of a cycle of adsorption and to compare these couples of adsorption.

Methodology: Five couples were considered: Silica gel-water, Zeolite-water, Activated Carbon-methanol, Activated Carbon-ethanol, and Activated Carbon-Ammonia.

From a thermodynamic cycle, heats exchanged was determined using Dubinin-Astakhov equation for adsorption and a COP is calculated.

We studied the influence of some physical quantities on the performance of these couples mainly the effect of operating temperatures and the effect of parameter n of Dubinin-Astakhov.

Results: The results showed that the silica gel-water and zeolite-water couples show better performance for evaporation temperature higher than 5 °C in the climatic conditions of Burkina Faso. On the opposite, for evaporation temperatures lower than 5 °C the activated carbon-methanol and activated carbon-ethanol couples have the best thermal performance. The results show that for each couple, there is an optimal value of Dubinin Astakhov parameter *n*:

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For silica gel-water n = 1.78, for zeolite-water n = 2.3, for activated carbon-methanol n = 2.15, for activated carbon-ethanol n = 2.25 and for activated carbon-ammoniac n = 1.55.

Finding(s): An interesting and useful finding was that the proposed thermodynamic model allow calculation of the COP of others refrigeration pairs knowing their thermophysicals properties and Dubinin Parameters.

We also found that silica gel-water and zeolite-water pair although not reliable in ice making purpose have higher cold production due to the high latent heat of vaporization of water

Conclusion: Silica gel-water pair is recommendable for refrigeration (food conservation) and activated carbon-methanol for ice making (Vaccine conservation).

The various results which are obtained by simulation need to be validated experimentally.

Keywords: Adsorbent/adsorbate; coefficient; energy; modelisation; performance; refrigeration; thermal.

NOMENCLATURES

Tc2	: Adsorption starting temperature [K]
Та	: Adsorption temperature [K]
β	: Affinity coefficient
Q_2	: Chaleur sensible heat of the metal parts [kJ]
E_0	: Characteristic energy of adsorption [kJ]
COP_s	: Coefficient of Solar Performance
COP_{th}	: Coefficient of Thermal Performance
Тс	: temperature [K]
D	: Constant that characterizes the adsorbent / adsorbate couple
R	: Constants of gases [J.kg ⁻¹ .K ¹]
e	: Criterion of Convergence
Δm	: Cycled mass of the adsorbate [kg]
$r_{l}(T)$: Density of the adsorbate [kg.m ³]
Q_{des}	: Desorption heat [kJ]
Tc1	: Desorption starting temperature [K]
$P_e(T)$: Evaporation pressure [Pa]
Te	: Evaporation temperature [K]
n	: Exponent in the Dubinin equation
Q_f	: Heat exchanged at the evaporator [kJ]
V ₀	: Maximum volume of adsorption per kg of adsorbent $[m^3.kg^{-1}]$
Tg	: Regeneration temperature [K]
$P_s(T)$: Saturation pressure of the adsorbate at temperature T [Pa]
Q_1	: Sensible heat of the adsorbent [kJ]
Q_3	: Sensible heat of the adsorbent [kJ]
Cp_a	: Specific heat of the solid adsorbent $[kJ.kg^{-1}.K^{-1}]$
V	: Volume adsorbed per kilogram of adsorbent [m³.kg ⁻¹]

INDICES

Ac	: Activated Carbon
а	: Adsorbent
Ads	: Adsorption
Amo	: Amoniac
С	: Condensation
Des	: Desorption
Eth	: Ethanol
е	: Evaporation
g	: Generation
lsos	: Isosteric
max	: Maximum
Meth	: Methanol
min	: Minimum
Sili	: Silicagel
Wat	: Water
Zeol	: Zeolite

1. INTRODUCTION

Nowadays, the world is in a deep energy crisis related to oil. Moreover, the environmental problems in the domain of refrigeration working with gas, solar thermal conversion, and particularly the adsorption refrigeration machines, meet renewed interest [1].

In developing countries this type of device can constitute an interesting sector for the producing of cold from solar energy.

Indeed, Burkina Faso for instance, a country located in the Sahel, has a very important solar potential with an average irradiation between 5.5 kWh.m⁻².day⁻¹ and 6.5 kWh.m⁻².day⁻¹ and an annual sunshine duration of 3000 hours running up to 3500 hours, with many rural areas that are not connected to the national electricity grid [2]. The use of solar adsorption refrigeration machines will undoubtedly improve the living conditions in these areas.

Several small prototypes were made with different adsorption couples both in laboratory and in situ under different climates [3–10] have demonstrated the usefulness of these machines.

Several authors have used the theories of Polanyi and Dubinin-Astakov. These seem to be the best theories available for the description of adsorption cycles [11].

A good evaluation of this theory is given by Luo et al. [12] with many simplifications.

The study carried out by [3] resulted in an approximate analytical formulation of the

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coefficient of performance of solar refrigerators with solid adsorption, depending on the physicochemical characteristics of the couple of adsorption and the particular conditions of use by limiting the scope to the couples 13x zeolitewater and silica gel-water only.

Pons et al. [13] conducted a comparative study on the three refrigeration systems (liquid absorption, adsorption with chemical reaction and adsorption) and concluded that the adsorption systems, although they have slightly lower returns than liquid absorption, present more advantages for the fact that they do not require the use of a pump.

In order to improve the COP of refrigeration with adsorption, several models of adsorption cycles have been proposed in the literature, generally with one or two couples of adsorption [14–22].

The study carried out by Allouhi et al. [23] on seven adsorption couples, by focusing on the adsorption capacity and the Solar Coefficient of Performance (COPs) lead to the following rank order of COPs: Silica gel-water, Fibrous activated carbon-methanol, Activated carbonmethanol, Zeolite-water, Activated carbonethanol, Activated carbon - ammoniac, Zeoliteethanol.

Experimental studies of adsorption refrigeration machines showing the transient operation of each component of the machine, carried out by other authors [24–34] showed that the COPs is generally between 0.05 and 0.2 and concluded that these systems are well suited to the Sahelian climate.

Mathematical simulation models have been developed in the literature in order to optimize the performance of adsorption couples [35–37].

Various couples are used, but it is rare to find in the literature comparisons of adsorbentadsorbate couples of the same amount and under the same conditions. In general, studies made on this type of machines are comprehensive studies aimed at obtaining global average performances [38]. However, the choice of couple to use is essential when deciding to produce an adsorption machine. That is the reason why this study is performed to help select the couple, according to the particular type of application in Burkina Faso.

Currently, research focuses on improving the heat transfer properties of materials on the one

hand, and on the other, on the consolidated adsorbents, and a literature review on the subject is provided by [39].

Therefore, most authors agree on couples such as silica gel-water, zeolite-water for cooling; [40,41], and couples of activated carbon-alcohol for the producing of ice [42].

The specificity of the present work lies in the fact of comparing the COP_{th} of several adsorption couples for the same quantities of adsorbent and adsorbate.

The objective of this work is to develop a thermodynamic model that applies to several couples, for determining the COP_{th} of a cycle of adsorption and discuss the influence of physical parameters and the Dubinin parameter **n** on the performance of cycles.

2. MATERIALS AND METHODOLOGY

2.1 Principle of Adsorption

Adsorption is a surface phenomenon involving a solid adsorbent which has a large surface area per mass unit (several tens of m² per gram) and a substance that adsorbs called adsorbate. The surface of these solids contains attraction sites for the adsorbed molecules. Many porous materials such as activated carbon, silica gels and zeolites satisfy this condition.

2.2 Theory of Polanyi

The theory of potential was formulated and verified by Polanyi et al. [43,44].

According to the theory of Polanyi, a potential E exists on the surface of the adsorbent and adsorbed vapours exist in the compressed state by attractive forces acting on the surface at a distance from the surrounding space [45].

Potential E may be expressed by the equation of isothermal compression of working pressure from pressure P to pressure P_s (saturation pressure):

$$E = \int_{P}^{P_s} V dP = RT \ln \frac{P_s}{P} \tag{1}$$

Volume V occupied by the adsorbed phase according to the Dubinin Radushkevish theory is

connected to the porous volume V_0 and potential E by the following relationship:

$$V = V_0 \exp\left[-\left(\frac{E}{\beta E_0}\right)^2\right]$$
(2)

Where E_0 is the characteristic energy of adsorption; and β is the affinity coefficient characterizing the polarization of the adsorbent.

2.3 Dubinin Theory

By substituting equation eq1 in eq.2, we get the equation of gas adsorption isotherm on a microporous solid:

$$V = V_0 \exp\left[-D\left(T\ln\frac{P_s}{P}\right)^2\right]$$
(3)

Where $D = \frac{R^2}{\beta^2 E_0^2}$ is a constant which

characterizes the couple adsorbent / adsorbate.

Therefore, the adsorbed mass is given by:

$$m = V_0 \rho(T) \exp\left[-D\left(T \ln \frac{P_s(T)}{P}\right)^2\right]$$
(4)

Dubinin and Astakhov generalized the relation (Eq.4) by introducing an exponent n that is variable, which is used to describe adsorption by zeolites and other adsorbents like silica gel; [46,47].

This new equation called Dubinin-Astakhov (DA) is written in its general form as:

$$m = V_0 \rho(T) \exp\left[-D\left(T \ln \frac{P_s(T)}{P}\right)^n\right]$$
(5)

2.4 Modeling of the Thermodynamics Cycle of Adsorption

An ideal adsorption cycle consists in four thermodynamic phases that can be represented with the Clapeyron diagram as shown in Fig. 1.

Isosteric heating phase (constant adsorbed mass)

- Isobaric desorption phase (constant pressure)
- Isosteric cooling phase,
- Isobaric adsorption phase.

The definition of the thermodynamic cycle of an adsorption machine requires the determination of the temperature of the two critical points Tc_1 and Tc_2 (thresholds).

The threshold desorption temperature Tc_1 which is the minimum temperature from which the phenomenon of desorption begins, corresponds to the appearance of the first drop of liquid in the condenser.

The threshold adsorption temperature Tc_2 is defined as the temperature at which the adsorption begins; it corresponds to the evaporation of the first liquid drop in the evaporator.

An analytical relationship between these threshold temperatures can be obtained by considering the two isosteric (A \rightarrow B) and (C \rightarrow D) in Fig. 1.



Fig. 1. Clapeyron diagramme of an ideal cycle

A prototype example is show in Fig. 2 which is a solar fridge with silica gel-water couple built in Ouagadougou.

We can write for Isosteres AB and CD respectively:

$$m(T_a, P_e) = m(Tc_1, P_c)$$
(6)

And

$$m(T_g, P_c) = m(Tc_2, P_e)$$
(7)

By applying equation (5) to equations (6) and (7) we respectively get:

$$\rho_{i}(T_{a})\exp\left[-D\left(T_{a}\ln\frac{P_{s}(T_{a})}{P_{e}}\right)^{n}\right] - \rho_{i}(T_{c1})\exp\left[-D\left(T_{c1}\ln\frac{P_{s}(T_{c1})}{P_{c}}\right)^{n}\right] = 0$$
(8)

$$\rho_{l}(T_{g})\exp\left[-D\left(T_{g}\ln\frac{P_{s}(T_{g})}{P_{c}}\right)^{n}\right] - \rho_{l}(T_{c2})\exp\left[-D\left(T_{c2}\ln\frac{P_{s}(T_{c2})}{P_{e}}\right)^{n}\right] = 0$$
(9)

Equation 8 and equation 9 are non-linear and the values of Tc_1 and Tc_2 are obtained by root finding using Newton (–Raphson) method.

Newton's method for solving $f(T_{c1}) = 0$ draws the tangent to the graph of $f(T_{c1})$ at any point and determines where the tangent intersects the x-axis. The method requires one starting value T_{c120} .



Fig. 2. Photo of the solar refrigerator with silica gel-water couple installed in Ouagadougou

2.5 Coefficients of Performance

The model developed is a static model, based on the use of equations of state (adsorbent / adsorbate) at thermodynamic equilibrium. It takes no account of the effects of adsorption kinetics, or the gradients of temperature or pressure caused by heat and mass transfer, and it is assumed that for a given variable, the other variables remain constant. Thus, we neglect the effect of thermal losses associated with the heating of the adsorber.

The efficiency of the machine is shown by the coefficient of thermal performance COP_{th} , this coefficient is useful for describing the operation of such a machine, and it is given by:

$$COP_{th} = \frac{Q_f}{Q_c} \tag{10}$$

Where Q_f is the production of cooling, the amount of cold produced at the evaporator.

 Q_c is the amount of heat supplied to the adsorber.

2.5.1 Determination of cold production Q_f

The heat exchanged in the evaporator Q_f is the difference between the heat extracted by evaporation of a mass Δm of the adsorbate at temperature T_e and the sensible heat corresponding to the refrigeration of the condensed cooling fluid from temperature T_c to temperature T_e of evaporation.

Hence

$$Q_f = m_a \Delta m \left[L(T_e) - \int_{T_e}^{T_c} Cp_l(T) dT \right]$$
(11)

L(T) and $Cp_l(T)$ respectively, denote the latent heat of evaporation and the specific heat of the adsorbate in liquid state.

 m_a is the mass of the solid adsorbent contained in the adsorber.

 Δm is the cycled mass of the adsorbate, calculated as follows:

$$\Delta m = m_{\text{max}} - m_{\text{min}} = m(T_a, P_e) - m(T_g, P_c) \quad (12)$$

 $m_{\rm max}$ is the adsorbed mass corresponding to temperature T_a of adsorption and evaporation pressure P_e , that is to say the adsorbed mass during the isosteric heating period.

 $m_{\rm min}$ is the adsorbed mass corresponding to temperature T_g of regeneration and the condensation pressure P_c , that is to say the adsorbed mass in the isosteric cooling.

2.5.2 Determination of the energy received by the adsorber $Q_{c.}$

Part of the energy received by the adsorber is used to heat the metal parts of the adsorber and another part is used to heat the adsorbent and the adsorbate and the rest is used for desorption. The energy received by adsorber Q_c is such that:

$$Q_c = Q_1 + Q_2 + Q_3 + Q_{des}$$
(13)

with:

- Sensible heat of the adsorbent:

$$Q_{1} = m_{a} \int_{T_{a}}^{T_{g}} Cp_{a} dT = m_{a} Cp_{a} (T_{g} - T_{a})$$
(14)

- Sensible heat of the metal parts:

$$Q_{2} = m_{g} \int_{T_{a}}^{T_{g}} Cp_{g} dT = m_{g} Cp_{g} (T_{g} - T_{a})$$
(15)

Where m_g is the mass of the metal parts of the adsorber and Q_a their specific heat.

- Sensible heat of the adsorbate Q_3

$$Q_{3} = m_{a}^{T_{a}} \int_{T_{a}}^{T_{a}} m(T) C p_{l}(T) dT = m_{a} m_{max} \int_{T_{a}}^{T_{a}} C p_{l}(T) dT + m_{a}^{T_{a}} \int_{T_{a}}^{T_{a}} m(T) C p_{l}(T) dT$$
(16)

m(T) is the mass adsorbed at temperature T and condensation pressure P_c (phase (B \rightarrow C) in Fig. 1).

- Heat of desorption Q_{des}

The desorption heat is given by Mimet (1991):

$$Q_{des} = m_a \int_{m_{max}}^{m_{min}} Q_{isost} dm$$
(17)

With $Q_{isost} = \Delta H_{ads}$

By differentiating the mass, we get:

$$dm = nDmT^{n} \left(\ln \frac{P_{s}(T)}{P} \right)^{n-1} \left[d \ln P - \frac{Q_{isost}}{RT^{2}} dT \right]$$
(18)

During this phase the pressure is constant and equal to the saturation pressure at the condensation temperature; therefore $d \ln P = 0$.

Expression of the heat of desorption is obtained by:

$$Q_{des} = m_a n D \int_{T_{c1}}^{T_s} m(T) T^n \left(\ln \frac{P_s(T)}{P_c} \right)^{n-1} \frac{Q_{isos}^2}{RT^2} dT$$
 (19)

The equation of heat received by the adsorber Q_c is thus expressed as:

$$Q_{c} = (m_{a}Cp_{2} + m_{g}Cp_{g})(T_{g} - T_{a}) + m_{a}m_{max}\int_{T_{a}}^{T_{c1}}Cp_{l}(T)dT + m_{a}nD\int_{T_{c1}}^{T_{s}}m(T)T^{n}\left(\ln\frac{P_{s}(T)}{P}\right)^{n-1}\frac{Q_{isout}^{2}}{RT^{2}}dT$$
(18)

2.6 Algorithm of Calculation

For the resolution of equations with this model, we write a Matlab code for calculating the threshold temperatures, calculating the various amounts of heat exchanged during the thermodynamic cycle and the thermal coefficient of performance.

The algorithm of this program is described in Fig. 3.

The parameters used in this simulation are shown in Tables 1 and 2.

The copper used as a building material of the adsorber has mass $m_g = 5 \ kg$ and its specific heat is $Cp_g = 0.380 \ \text{KJ/kg.K}$. The thermodynamic properties $P_s(T)$, $\rho(T)$, L(T) and $Q_1(T)$ of the fluids used are obtained by polynomial approximation of thermodynamic functions given by Polynomial data Temperature-Property Relationship [48].

Couple	V0 (l/kg)	D	n	Reference
Silicagel-water	0.350	6e-6	1.78	Simulation
AC35-CH₃OH Zeolite-water AC35-C₀H₅OH	0.425 0.269 0.439	5.02e-7 1.80e-7 1.60e-7	2.15 2.25 2.25	M.Pons, PH Grenier [5] J-J Guilleminot [49] Simulation
AC35-NH ₃	0.456	0.53e-4	1.49	Simulation

Table 1. Dubbinin asthakov Parameters of the different couples

Table 2. Mass and specific heat

	$Cp_a(KJ/Kg.K)$	$m_a(kg)$
Silicagel	0.837	1
Zeolithe	1.04	1
Activated Carbon	0.920	1



Fig. 3. Algorithm of calculation

3. RESULTS AND DISCUSSION

Depending on the thermodynamic model, the COP_{th} and the amount of cold produced in the evaporator have been calculated for the different couples by varying a parameter.

Fig. 4 shows the variation of the thermal coefficient of performance COP_{th} according to T_{g} .

We note that for the couples studied, COP_{th} increases with T_g up to a maximum reached for a recorded optimum temperature $T_{g\,max}$. For temperatures above this value the COP_{th} decreases. This behaviour can be justified by the fact that beyond a certain regeneration temperature, the heating energy is only used to increase the temperature of the adsorbent, the temperature of the metal parts of the adsorber and the temperature of the water, in spite of m_{min}

the mass of desorbed water, is becoming weaker [50].

Equation (13) indicates that the decrease $m_{\rm min}$ causes an increase in the cycled mass Δm ,

which leads to an increase of the amount of cold produced in the evaporator Q_f (Fig. 5) according to equation (12).



Fig. 4. Influence of the temperature of regeneration Tg on the COP_{th} (Ta=298 K,Te=278 K ,Tc=303 K)



Fig. 5. Influence of the regeneration temperature Tg on Q_f (Ta=298 K, Te=278 K, Tc=303 K)

Fig. 6 shows that the COP_{th} decreases with the increase of condensing temperature T_c .

Indeed, the increase of T_c results in an increase of the saturation pressure at this temperature $P_s(T_c)$. Therefore, the desorbed mass

 $m(T_g, P_s(T_c))$ decreases, implying a reduction in the cycled mass according to equation (12) and consequently, a decrease in the amount of cold produced in the evaporator Q_f (see Fig. 7) and the COP_{th} of the system.



Fig. 6. Influence of the condensation temperature Tc on the COP_{th} (Ta=303 K, Te=278 K, Tg=373 K)



Fig. 7. Influence of the condensation temperature Tc on $Q_{\!f}$ (Ta=303 K, Te=278 K, Tg=373 K)

Figs. 8 and 9 show respectively the variations of COP_{th} and the quantity of cold Q_f produced in the evaporator.

There is an increase in COP_{th} and in the quantity of cold produced in the evaporator when the temperature of evaporation increases.

Indeed, the increase in the evaporating temperature $T_{e}\,$ causes an increase in the

saturation pressure at this temperature $P_s(T_e)$. Therefore, the mass of water adsorbed $m(T_a, P_s(T_e))$ increases, involving an increase in the cycled mass given by equation (13) and therefore, an increase of the amount of cold produced at the evaporator Q_f and the COP_{th} of the system.









The evolution of parameters Q_f and COP_{th} with respect to the adsorption temperature are illustrated respectively by Figs. 10 and 11.

We observe that the increase in adsorption temperature results in a decrease in COP_{th} and Q_t .

This is explained by the fact that, with the relationship of the adsorbed mass according to the Dubinin Astakhov model, an increase in T_a

results in a decrease in adsorbed mass corresponding to that temperature and the saturation pressure at the evaporation temperature $m(T_a, P_s(T_e))$. Therefore the cycled mass decreases, which also results in decreased $_{COP_a}$ and Q_f .

The influence of parameter **n** of Dubinin Asthakov on the COP_{th} and Q_f is respectively illustrated on Figs. 12 and 13.



Fig. 10. Influence of adsorption temperature Ta on the COP_{th} (Tc=278 K, Tc=303 K, Tg=373 K)



(Tc=278K, Tc=303K, Tg=373K)

One can observed that for each couple, there is an optimal value of the parameter n of Dubinin Astakhov: Thus, for silica gel-water n = 1.78 for zeolite-water n = 2.3, for activated carbonmethanol n = 2.15, for activated carbon-ethanol n = 2.25, for activated carbon-ammonia n = 1.55.

The results show that silica gel-water and zeolitewater couples exhibit better performance for evaporation temperatures above 5°C in the climatic conditions of Burkina and are therefore suitable for the conservation of vaccines.

In the opposite, for evaporation temperatures lower than 5°C the activated carbon-methanol and activated carbon-ethanol couples have the best thermal performance. The latter's are indicated for the producing of ice or lowtemperature cooling.



Fig. 12. Influence of Dubinin-Astakhov parameter n on the *COP*_{th} (Ta=303 K, Te=278 K, Tc=303 K, Tg=373 K)



Fig. 13. Influence of Dubinin-Astakhov parameter n on Q_f (Ta=303 K, Te=278 K, Tc=303 K, Tg=373 K)

Adell [3] concluded that silica gel is substantially more efficient than zeolite as regards the temperature of use of cold greater than 0° C, by comparing the silica gel-water and water-zeolite couples.

4. CONCLUSION

The study of thermodynamic cycle performed in this work for five adsorption couples demonstrated the importance of an appropriate choice of a couple when we need a given application.

This is useful for the design of adsorption system.

The silica gel-water and zeolite-water working pair showed high performance for refrigeration cycles purpose while activated carbon-methanol and activated carbon-ethanol showed to be the most promising working pairs for ice making purpose.

These results showed that under conditions of simulation we can classify these couples: silica gel-water, zeolite- water, activated carbon-methanol, activated carbon-ethanol, and activated carbon -ammoniac.

The parameter n of Dubinin has a great influence on the modeling of adsorption and for every couple there is an optimal value.

Based on the simulation results we can recommend silica gel-water pair for refrigeration (food conservation) and activated carbonmethanol for ice making (Vaccine conservation). This approach could also be applied to other adsorbent/adsorbate pairs for the design of adsorption beds.

The various results which are obtained by simulation need to be validated by experimental methods.

A forthcoming publication will be devoted to an experimental comparative study of these couples.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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