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1 Experimental study of water evaporation rate, at the surface of
2 aqueous solution, under the effect of a discontinuity of chemical
3 potential - effect of water activity and air pressure

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12

13 **Abstract**

14 The aim of this communication is to analyse the influence of water activity and total pressure
15 on water evaporation. The system is composed of liquid and gas phases, separated by a plane
16 surface, contained in a cylinder whose volume is regulated by a piston. Water activity is
17 regulated by saturated salt solutions and pressure by the piston. The experimental device and
18 procedures were defined to limit the temperature variation at the interface. A transient
19 method is used. From a steady state, a volume increment is imposed; the resulting non-
20 equilibrium leads to an increase in the partial pressure of water vapour to the equilibrium
21 pressure imposed by the solution. Numerical calculation shows little variation in temperature
22 in the gas-liquid interface under the experimental conditions. An evaporation model is
23 adopted taking into account chemical potential discontinuity at the interface. The surface flux
24 evaporation and chemical potential jump at the interface are deduced from the total pressure
25 recording. In the neighbourhood of equilibrium, the surface flux of phase change is shown to
26 be proportional to the chemical potential jump. The surface coefficient of evaporation
27 increases with the total pressure of the gas phase and water activity.

28 **Key words:** surface evaporation rate; chemical potential; water activity; gas pressure;
29 transient method.

30 **Nomenclature**

31 a_w : Activity of water (/)

32 C_α : Specific heat capacity of the phases; $\alpha = g$ for the gas phase, $\alpha = l$ for the liquid
33 phase (J.kg⁻¹.K⁻¹)

- 34 D_v : Diffusion coefficient of water vapour in air ($\text{m}^2.\text{s}^{-1}$)
- 35 h_g : Thickness of the gas phase (m)
- 36 h_l : Thickness of the liquid phase (m)
- 37 J_v : Evaporation flux by surface unit and unit time ($\text{kg}.\text{m}^{-2}.\text{s}^{-1}$)
- 38 J_{di} : Diffusive flux of the vapour ($i=v$) and air ($i=a$) ($\text{kg}.\text{m}^{-2}.\text{s}^{-1}$)
- 39 L^S : Isothermal phase change coefficient by unit surface ($\text{kg}.\text{K}.\text{s}.\text{m}^{-4}$)
- 40 M : Molar mass of water ($0.018 \text{ kg}.\text{mol}^{-1}$)
- 41 p_g : Pressure of the gas phase (Pa)
- 42 p_i : Partial pressure of air, ($i=a$) and vapour ($i=v$) (Pa)
- 43 p_v^{eq} : Partial pressure of water vapour at the equilibrium with water in a solution (Pa).
- 44 $p_{vs}(T)$: Saturated vapour pressure at temperature (T)
- 45 p^+ : Reference pressure (Pa)
- 46 p_0 : Standard pressure (101325 Pa)
- 47 Q : Energy flux ($\text{J}.\text{m}^{-2}.\text{s}^{-1}$)
- 48 R : Perfect gas constant ($8.314 \text{ J}.\text{K}^{-1}.\text{mol}^{-1}$)
- 49 T : Temperature (K)
- 50 T_l : Temperature of the liquid phase (K)
- 51 T_v : Temperature of water vapour (K)
- 52 T_0 : Standard temperature (273.15 K)
- 53 v_α^k : Gas phase velocity ($\alpha = g$), of air ($\alpha = a$) and water vapour ($\alpha = v$) ($\text{m}.\text{s}^{-1}$)
- 54 z : Elevation from the liquid-gas interface (m)
- 55
- 56 *Greek symbols*
- 57

- 58 λ_α : Thermal conductivity of the phases; ($\alpha = g$) for the gas phase, ($\alpha = l$) for the liquid
59 phase ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)
- 60 μ_v : Mass chemical potential of water vapour ($\text{J}\cdot\text{kg}^{-1}$)
- 61 μ_w : Mass chemical potential of liquid water ($\text{J}\cdot\text{kg}^{-1}$)
- 62 μ^+ : Reference mass chemical potential reference, function of the temperature T , at
63 reference pressure P^+ ($\text{J}\cdot\text{kg}^{-1}$)
- 64 ρ_α : Mass density of gas phase ($\alpha = g$), of air ($\alpha = a$) and water vapour ($\alpha = v$) ($\text{kg}\cdot\text{m}^{-3}$)
65
- 66 ε : Correcting coefficient for evaporation rate (l)

67

68 1. Introduction

69 Liquid-gas phase change is involved in many mass and energy transfer. Examples include
70 evaporation over large bodies of water (lakes and seas) and their impact on climate,
71 evaporation from the soil surface associated with desertification, industrial distillation and
72 refining processes, high temperature geothermal processes, drying of food and industrial
73 products.

74 Surface evaporation involves mass and energy transfer phenomena related to:

- 75 (i) temperature discontinuity at the interface and its variations on both sides of the
76 interface,
- 77 (ii) convective and diffusive transport in both phases,
- 78 (iii) chemical potential discontinuity at the interface, generally associated with a difference
79 between the vapour partial pressure and the equilibrium vapour pressure of liquid
80 water.

81 The simultaneous consideration of all these mechanisms and their coupling makes theoretical
82 and experimental analysis difficult. Concerning phenomena (i) and (ii), Fang and Ward [1]
83 performed a phase change experiment for a spherical interface separating pure water and
84 water vapour. Partial pressure was kept constant at values between 194.7 and 596 Pa by a
85 flux of vapour. The tests were carried out for temperatures of 15, 26 and 35°C. The average
86 surface evaporation flux varied between 70 $\mu\text{l}/\text{h}$ and 160 $\mu\text{l}/\text{h}$. The temperature was measured
87 in the fluid and gas phases by thermocouples. The measurements showed an interfacial
88 temperature discontinuity up to 8°C for evaporation rates of 160 $\mu\text{l}/\text{h}$. The temperature on the
89 vapour side was greater than the temperature on the liquid side.

90 Concerning phenomena (i) and (ii), Gatapova et al. [2] brought heat locally to a thin layer of
91 water at atmospheric pressure. Temperature and heat flux are recorded during experiments.
92 Various thermocouples (4 μm diameter) allow measuring the temperature jump at the water-
93 air interface. A temperature jump of about 0.2°C is detected for the liquid surface temperature
94 of 35.2°C and the heater temperature of 43.6°C. They conclude that the value of temperature

95 jump depends not only on evaporation intensity but also on the temperature gradients in
96 liquid and gas phases near the interface.

97 Kazemi et al. [3] conducted an experimental and theoretical study on the non-stationary
98 evaporation of water at low pressure. They proposed a mathematical model of the system
99 which takes into account mass, momentum and heat transport in liquid and gas phases, as
100 well as the phase change at the interface. The study allowed the description of the mass
101 transfer through the interface. The experiments consisted in measuring the velocity field in
102 the central plane of the evaporating water. In addition, the temperature distribution at the
103 liquid-vapour interface was measured with a fine thermocouple. The results showed good
104 correlation between simulation and experiment. So, this model was used to reveal certain
105 behaviours on the phenomenon of evaporation difficult to access by experiments alone.

106 In the experiments proposed by Badam et al. [4], concerning phenomena (i), a cylindrical
107 water vapour interface is subjected to heat input. Heaters provided energy to the interface. A
108 moving thermocouple was used to measure the temperature profile in the liquid and vapour
109 phases. The water flow at the interface was determined from the quantity of water injected
110 into the liquid phase. These measurements were used to deduce the flow of evaporated water
111 and the temperature discontinuity at the interface, as well as heat flows in the liquid and gas
112 phases near the interface. Temperature discontinuity depended on the imposed energy; it was
113 about 2°C without input of heat energy, and reached values up to 10°C when the temperature
114 was regulated at 80°C. These values are approximately 10 to 20 times higher than those given
115 by the kinetic theory of gases [4].

116 The experiments mentioned that the addition or removal of mass or heat at the interface
117 increases phase change. The release of latent heat of cools the two phases and leads to
118 temperature discontinuity at the interface. These experiments are performed in steady state,
119 the temperature jump is determined at the end of the experiment, which supposes that the
120 interface has reached an equilibrium, which is far from immediate.

121 In the environment, solvent, usually water, is generally impure. When the water contains salts
122 or other chemical compounds in solution, the activity of the water is modified as well as the
123 evaporation rate. Limited to aqueous saline saturated solutions, the objective of the
124 experimental study proposed here is to validate a macroscopic evaporation law triggered by a
125 chemical potential at the interface (phenomenon (iii)) [4, 5, 6]. The influence of aqueous water
126 activity and total gas pressure on the evaporation rate will be analysed in this contribution.

127

128 **2. Experimental studies of gas-liquid evaporation with low temperature** 129 **variation**

130 The choice of a jump in chemical potential as the cause of evaporation requires carrying out
131 an experiment that minimizes the magnitude of mechanisms (i) and (ii); this is achieved if the
132 experiment is carried out in a closed environment in the absence of diffusion and if the
133 temperature variation at the interface is very low. However, the imposed conditions must be
134 able to create an interfacial discontinuity of the chemical potential. To avoid a prevailing
135 temperature jump at the interface and to restrict the movement of fluids, the experiments are
136 carried out under transient conditions. In a close cylinder containing liquid and vapour water,

137 the phase change will be triggered by a rapid supply of mechanical energy. This energy will
138 be provided by an increment of volume occupied by the gaseous phase, causing the
139 vaporization of a small quantity of water. Physical quantities will be recorded during the
140 return to equilibrium in order to analyse the phase change resulting from a discontinuity of
141 the chemical potential. However, this experimental procedure can still induce evaporation
142 under the action of mechanisms (i) and (ii), requiring to discuss the assumptions made.
143

144 *Description of the experimental device*

145 The experimental device, presented in Fig.1, was recently developed to characterize the
146 liquid-gas equilibrium in hygroscopic porous materials [7]. The same apparatus is used here
147 to analyse the kinetics between two equilibrium situations.

148 Water activity is controlled by means of standard saturated salt solutions (Tab.2). The
149 solution under investigation was placed in a cylindrical tank (a), its thickness being of the
150 order of 5 mm. A piston pump (c) was placed above the solution in order to apply gas
151 pressure in (g) below the atmospheric pressure. The piston chamber volume is modified by a
152 screw system (d) with a graduated ruler (e). O-ring gaskets (f) ensured perfect air tightness so
153 that the system could be considered closed.

154 **Figure 1**

155 **Table 1**

156 Some characteristic dimensions are given Tab.1. A pressure transducer (b) (Druck,
157 PMP4030AB) was used to record the total pressure of the gas phase, p_g , throughout the
158 process with accuracy of ± 10 Pa. The whole device was placed in a thermo-regulated bath at
159 30°C to ensure a constant temperature at the outer surface of the cylinder. As the device was
160 made of stainless steel its high thermal inertia led to low temperature variations. Data
161 acquisition was performed through a National Instrument DAQ card and analysed using
162 LabView software.

163 *Operating mode*

164
165 The tests were carried out for total pressures of 55, 75 and 95 kPa. In order to regulate both
166 the total pressure of the gas phase and the partial pressure of water vapour, the following
procedure was adopted.

167 1/ To adjust the final total pressure, preliminary experiments were carried out. The solutions
168 were introduced and the piston was placed in the low position. By using the pressure sensor,
169 the total pressure was brought to the desired value. The device was then disassembled and the
170 height of the cavity (g), noted h_g was measured. The gaseous phase being composed of 96%
171 of air, in the most unfavourable case of pure water, h_g depended little on the composition of
172 the solution. For the three pressures (55, 75 and 95 kPa), the average values of h_g were
173 respectively: 14.22, 24.09 and 26.24 mm.

174 2/ As can be seen in Tab.2 the minimum equilibrium partial pressure was 500 Pa for LiCl. It
175 was decided to impose a partial pressure variation of 500 Pa with respect to the equilibrium

