



# Antibody-Gold Nanoparticle Bioconjugates for Biosensors: Synthesis, Characterization and Selected Applications

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

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## Abstract

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

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

## Nomenclature

$a_w$  : Activity of water (/)

$C_\alpha$  : Specific heat capacity of the phases;  $\alpha = g$  for the gas phase,  $\alpha = l$  for the liquid phase (J.kg<sup>-1</sup>.K<sup>-1</sup>)

- 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  $v_\alpha^k$  : 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	$\lambda_\alpha$	: Thermal conductivity of the phases; ( $\alpha = g$ ) for the gas phase, ( $\alpha = l$ ) for the liquid
59		phase ( $\text{W.m}^{-1}.\text{K}^{-1}$ )
60	$\mu_v$	: Mass chemical potential of water vapour ( $\text{J.kg}^{-1}$ )
61	$\mu_w$	: Mass chemical potential of liquid water ( $\text{J.kg}^{-1}$ )
62	$\mu^+$	: Reference mass chemical potential reference, function of the temperature $T$ , at
63		reference pressure $P^+$ ( $\text{J.kg}^{-1}$ )
64	$\rho_\alpha$	: Mass density of gas phase ( $\alpha = g$ ), of air ( $\alpha = a$ ) and water vapour ( $\alpha = v$ ) ( $\text{kg.m}^{-3}$ )
65		
66	$\varepsilon$	: Correcting coefficient for evaporation rate ( $/$ )

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  $\mu\text{l/h}$  and 160  $\mu\text{l/h}$ . 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  $\mu\text{l/h}$ . 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  $\mu\text{m}$  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

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

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

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

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

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

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

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

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

### ***Description of the experimental device***

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

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

### **Figure 1**

### **Table 1**

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

### ***Operating mode***

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

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

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

