

# FUNDAMENTALS OF MOISTURE AND ENERGY FLOW IN CAPILLARY-POROUS BUILDING

## MATERIALS

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## ABSTRACT

Basic physical features of combined energy and moisture flow in porous building materials for dynamic and stationary processes are discussed. The mathematical and physical structure of these dynamic processes is discussed in terms of local thermodynamical equilibrium, flows induced by gradients in intensive state variables, and conservation of energy, moisture and other components.

The conditions for thermodynamical equilibrium in a capillary-porous material containing moisture in liquid and gas phases, air, and possibly salts in the pores are presented as a prerequisite for the understanding of the dynamic flow processes. The peculiarities of pore-water tension are discussed. Moisture may flow in gas phases and in liquid phases while interacting in a complicated way. The specific problems arising from the complex character of the flow are discussed. The relation between the moisture flow and gradients in intensive state variables such as pore-water pressure or tension, vapor pressure, gas pressure and temperature is discussed.

Some problems concerning the energy flow are discussed. The energy flow is complicated by the mass flow in the pores and especially by continuous condensation and evaporation of water in the pores. Divisions of the energy flow into heat flow and energy transfer due to mass flow and the effects of condensation and evaporation are discussed.

The equilibrium relations and the transfer coefficients for porous materials often exhibit rather pronounced hysteresis. The causes of hysteresis and the complications due to hysteresis in the description, are dealt with.

## INTRODUCTION

A building material like concrete or brick is penetrated by a complex pore system. The pore space is filled by air, water and possibly salts. Suitable parts of the pore volume are occupied by capillary condensed liquid water. The free pore walls are covered by layers of adsorbed water molecules. The remaining parts of the pore space contain a mixture of air and water-vapour. The phases of liquid water may contain salts.

Consider a small part or region of the porous material. The region is small compared with the overall dimensions of the considered piece of building material, but it is large compared with the minute single pores. The region contains a reasonably representative sample of the pore structure. The state of such a region is characterized by a number of variables like the moisture

content  $w$  ( $\text{kg H}_2\text{O}/\text{m}^3$ ), the temperature  $T$ , the water-vapour pressure  $p$ , and so on. We will consider such a region as a point.

The state will usually vary from point to point in the material. The distribution of states in the material will change with the time  $t$  during a dynamical process when moisture and energy flow through the porous material. The aim of the study of these processes is to be able to calculate how this distribution of states through the building material evolves with time. The states at an initial time and the conditions at the boundaries during the dynamical process have to be known.

## MATHEMATICAL STRUCTURE

The mathematical description is based on the conservation equations for moisture and energy. We will, for the sake of notational simplicity, consider the one-dimensional case with flows and changes only in the direction of  $x$ . Let  $F_w$  and  $F_e$  denote the flows of moisture and energy respectively. The moisture flow  $F_w(x,t)$  in a point  $x$  at a time  $t$  refers to the total mass of the water molecules that pass the point  $x$  per unit time and unit area perpendicular to the flow direction  $x$ . The moisture flow  $F_w$  may have contributions from liquid flow, convective and diffusional gas flow and possibly migrations along the pore walls. The energy flow  $F_e$  refers to the total energy flow. The heat flow  $F_q$  is one contribution to  $F_e$ .

The moisture content, i.e. the total mass of water molecules in the myriads of liquid, gas and surface phases per unit volume of the porous material, is  $w$ . Let  $e$  denote the total energy content per unit volume. (It is preferable to use moisture and energy contents per unit mass of dry solid in situations where volume changes of the solid are of importance.)

The introduced quantities  $F_w$ ,  $F_e$ ,  $w$  and  $e$  are functions of  $x$  and  $t$ . The equations for conservation of moisture and energy are:

$$\frac{\partial w}{\partial t} = -\frac{\partial F_w}{\partial x} \qquad \frac{\partial e}{\partial t} = -\frac{\partial F_e}{\partial x} \qquad (1)$$

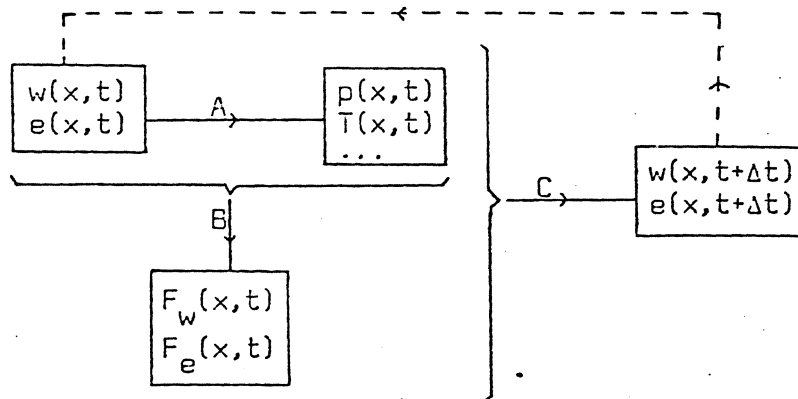
The left-hand members give the increase in moisture and energy content in a point (per unit time and unit volume). The right-hand members give the net influx to a point (per unit time and unit volume) of moisture and energy respectively. The net influx depends on the rate of change of the flow intensity along the  $x$ -axis.

We must now have suitable expressions for the flows  $F_w$  and  $F_e$  in order to be able to perform the calculations. These expressions serve to relate the flows to the present distribution of states through the material. The flows present a paramount problem. The flows in a point  $x$  at a time  $t$  depend on the present state at the point and on the slightly differing states in the vicinity of the point. Differently phrased, the flows depend on present gradients of state variables at the point and on the state at the point.

We must be able to specify the flows that arise for every distribution of moisture and energy that occurs during the dynamical process. The conservation equations (1) express, once one has got used to them, a rather trivial book-keeping for moisture and energy. The difficulties and the main physical content lie in the actual expressions for the flows, in which these flows are related to the spacial rates of change and to the levels of various state variables.

It is often a bit difficult to discern the structure behind the great variety of alternative, more or less clearly formulated, conservation equations that are used. However, the mathematical structure contained in the conservation equations (1) is indeed simple. Suppose that the moisture and energy distributions  $w(x,t)$  and  $e(x,t)$  are known at a time  $t$ . Then the state is known at each point. A certain information concerning the preceding moisture history

for each point may also be necessary due to hysteresis effects. The complete knowledge of the present state in a point also requires a number of formulas that relate pertinent state variables like the temperature  $T$ , vapour pressure  $p$ , and so on to the conservation variables  $w$  and  $e$ . The expressions for the flows then give  $F_w(x,t)$  and  $F_e(x,t)$  for each point  $x$  at the considered time  $t$ . There must be additional expressions that render it possible to specify the flows through the boundaries during the dynamical process. From formulas (1) we obtain the rate of change of  $w$  and  $e$  for each point. From this we get  $w(x,t+\Delta t)$  and  $e(x,t+\Delta t)$  for each point  $x$  at a slightly later time  $t+\Delta t$ . The procedure is repeated for the new time  $t+\Delta t$ , and so on. This mathematical structure of the calculations is illustrated in figure 1.



A: Equilibrium relations    B: Expressions for the flows  
C: Conservation equations

Figure 1. Mathematical structure of a dynamical process with moisture and energy flow in a porous building material.

A numerical simulation of a dynamical process may closely follow the procedure of figure 1. The continuous moisture and energy distributions are approximated by the values in a number of discrete points or cells. A set of equilibrium relations allows the calculation of other pertinent state variables for each cell. The flows of moisture and energy through the boundary between two consecutive cells are calculated from the differences in the state variables between the two cells and from the prevailing levels of the state variables with the aid of the expressions for the flows. The boundaries require additional formulas that give the flows through these. Direct moisture and energy balances then give new moisture and energy contents for each cell after a time increment  $\Delta t$ . This procedure is repeated for time-step after time-step.

Differences in concentrations of other substances and changes due to flows of these substances may be of importance in the dynamical process. There is an additional conservation equation for each new substance. Consider as an example a case when the air and a salt in the pores are of importance. Let  $F_a$  (kg air/m<sup>2</sup> s) denote the flow of air and  $a$  (kg air/m<sup>3</sup>) the air content. Let  $F_b$  and  $b$  be the corresponding quantities for the salt. Then we get two additional conservation equations for the air ( $a$ ) and the salt ( $b$ ):

$$\frac{\partial a}{\partial t} = -\frac{\partial F_a}{\partial x} \qquad \frac{\partial b}{\partial t} = -\frac{\partial F_b}{\partial x} \qquad (2)$$

The difficult problem is again the expressions that make it possible to calculate the four flows, when the state variables are known through the porous material.

## THERMODYNAMICAL EQUILIBRIUM

It is of fundamental importance to base the studies of these dynamical processes on a thorough thermodynamical analysis of the behaviour of moisture and other substances in the pore system of a building material. The thermodynamics of the pore system with its different constituents provides the basis for all deeper physical insight in these processes. This thermodynamical system is in a way very complicated due to the complex pore structure in which liquid and gas phases are intertwined in a way that may seem completely intractable. However, the analysis of the conditions for thermodynamical equilibrium produces an amazingly large amount of valuable and useful information. The thermodynamical investigation also provides valuable insight into the behaviour in non-equilibrium, since we obtain the direction of the local flows. The intensities of the overall flows are given from measured or inferred transport and transfer coefficients.

Let us now consider a piece of a porous material in thermodynamical equilibrium. The pore space is occupied by liquid water phases and gas phases. We will always have a connected region in mind when the word phase is used.

A gas phase contains a mixture of air and water-vapour. All gas phases will in equilibrium be in exactly the same state. A gas phase deep inside the material will be in the same state as the surrounding humid air, even if these two phases are separated from each other by a long sequence of other liquid and gas phases. Thus all gas phases have the same gas pressure  $p_g$  and the same water-vapour pressure  $p$ . Consider as an illustration a non-equilibrium situation where a single gas bubble deep inside the material has a higher pressure  $p_g$  than that of the outside surrounding air. Air from the bubble will dissolve in neighbouring liquid phases and diffuse through the liquid out to the surrounding air. The gas bubble will eventually adjust its pressure to the equilibrium value or, if this is not feasible, vanish completely.

Let us first consider the case when the liquid phases do not contain any salts. Then all liquid water phases must in equilibrium be in exactly the same state. They have the same pressure  $p_l$ . The pore-water pressure  $p_l$  is in general different from the pressure  $p_g$  in the gas phases in the pores. The liquid and gas phases exchange water molecules. The chemical potential for water in the gas phases and in the liquid phases must in equilibrium be the same. This condition gives the so called Kelvin equation [1]. A very accurate expression for the condition of equal chemical potential is [2]:

$$p_l - p_s(T) = \frac{RT}{v_l} \ln \left( \frac{p}{p_s(T)} \right) \quad (3)$$

Here  $p_s(T)$  is the saturation vapour pressure of (pure) water at the temperature  $T$ . The temperature  $T$  is in degrees Kelvin, and  $R$  is the general gas constant. The quantity  $v_l$  is the volume of one mole of liquid water. The factor  $RT/v_l$  is equal to  $1340 \cdot 10^5$  Pa at room temperature. Table I shows corresponding values of relative humidity  $p/p_s$  and pore-water pressure  $p_l$  for  $T=293\text{K}$ .

$\frac{p}{p_s}$	0.8	0.9	0.99	0.999	1	1.001
$p_l$ (bar)	-300	-140	-13	-1.3	+0.02	+1

Table 1. Corresponding values of relative humidity  $p/p_s$  and pore-water pressure  $p_l$  at room temperature in a case without salts. (1 bar= $10^5$  Pa)

There are two conspicuous facts concerning the pore-water pressure  $p_l$ . The first is that  $p_l$  usually turns out to be negative. This means that there is a state of tension in the liquid phase. The second remarkable fact is the large numerical values of  $p_l$ . A change  $\Delta(p/p_s)=0.01$  in relative humidity corresponds to a change  $\Delta p_l=13 \cdot 10^5$  Pa in pore-water pressure or tension.

Equation (3) gives the condition for diffusional equilibrium between a liquid water phase and neighbouring water-vapour in a gas phase. Consider now a non-equilibrium situation with fixed relative humidity  $p/p_s$ . Water will evaporate from a liquid phase, if the pore-water pressure is higher than the equilibrium value according to equation (3). Conversely, water-vapour will condense on the liquid surfaces, if the pore-water pressure lies below the equilibrium value. Suppose as an example that the relative humidity is lowered from saturation  $p/p_s=1$  to  $p/p_s=0.90$ . Then, according to table 1, the pore-water pressure must fall to  $p_l=-130 \cdot 10^5$  Pa in order to prevent evaporation and maintain equilibrium. All liquid water will eventually evaporate if the liquid phase fails to establish this high tension or negative pressure. We know that there is liquid water in many porous materials in equilibrium below saturation. This means that there must exist a mechanism which has the ability to create and maintain these high tensions or negative pressures in the pore water.

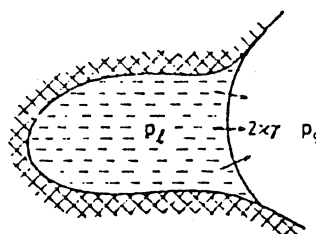
These states of high tension are caused by the surface tension of suitably curved water surfaces. Consider a point on a water surface in a pore. Let  $\kappa$  denote the so-called mean curvature of the surface in the point. The inverse of  $\kappa$  gives a mean radius of curvature. It can be shown with the aid of differential geometry [2] that the local net effect of the surface tension  $\gamma$  of the curved water surface is a force in the normal direction to the surface. The magnitude of the net force per unit area of water surface is  $2\kappa\gamma$ . This is an exact expression irrespective of the shape of the water surface. There is a pressure  $p_g$  in the gas phase outside the liquid and a pressure  $p_l$  in a liquid phase. The pressure difference  $p_g-p_l$  is balanced by the force  $2\kappa\gamma$ . This equation for force balance at a point on a curved water surface is due to Laplace:

$$p_g - p_l = 2\kappa\gamma \quad (4)$$

The force equilibrium is illustrated in figure 2. The mean curvature  $\kappa$  must in equilibrium have the same value at all points of all water surfaces in the pores, since  $p_g$  and  $p_l$  have the same values throughout. This is a strong condition on the shapes of the water surfaces.

Coercive forces between the water molecules keep the liquid phase together and prevent rupture. Coercive forces between the pore wall and the water in the vicinity of the walls keep the liquid attached to the pore walls in spite of the tension in the liquid.

Figure 2. Cross-section through a pore. The pressure difference  $p_g-p_l$  over the water surface is balanced by the force  $2\kappa\gamma$  from the surface tension  $\gamma$  of the curved water surface with mean curvature  $\kappa$ .



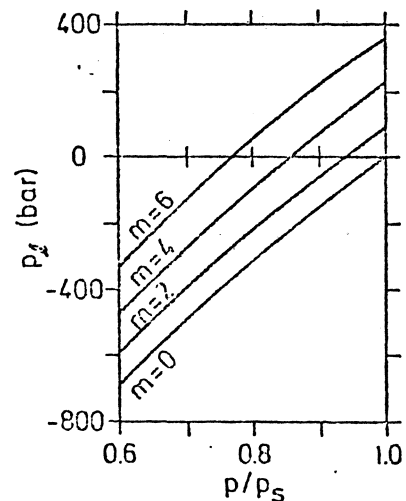
High pore-water tensions require heavily curved water surfaces. Take as an example a situation with  $p_\ell = -130 \cdot 10^5$  Pa, which corresponds to  $p/p_s = 0.90$ . The so-called mean curvature  $\kappa$  in any point at a free surface of a liquid phase is given by (4). The corresponding mean radius of curvature  $R = 1/\kappa$  becomes  $R = 110 \text{ \AA}$  ( $1.1 \cdot 10^{-8} \text{ m}$ ).

The liquid phases in the pores of a building material usually contain salts. These influence the chemical equilibrium condition between liquid water and water-vapour considerably. There remains a lot of research to be done concerning the influence of salts on the dynamical flow processes. The condition for equilibrium is now instead of (3):

$$p_\ell - p_s(T) = \frac{RT}{v_\ell} \ln \left( \frac{p}{p_s(T)} \right) + \frac{RT M_w}{v_\ell} v \phi m \quad (5)$$

The difference from (3) is the last term, which is of osmotic character. Here  $M_w$  is the mole weight of water,  $v$  the number of ions per dissolved salt molecule, and  $m$  the molality (i.e. the number of moles of salt per kilogram of water) of the salt in the considered liquid phase. The factor  $\phi$  is the molal osmotic coefficient for the salt dissolved in water [3]. It is a function of the concentration  $m$ . A reasonable approximation is usually  $\phi \approx 1$ . Relation (5) is shown in figure 3 for sodium chloride. We note as a rule of thumb that a decrease by ten per cent in relative humidity ( $\Delta(p/p_s) = -0.10$ ) or in relative weight of salt in the salt solution ( $\Delta m = -2$ ) corresponds to a decrease of pore-water pressure  $p_\ell$  of the order of  $100 \cdot 10^5$  Pa.

Figure 3. Relation between pore water pressure  $p_\ell$  and relative humidity  $p/p_s$  for different molalities  $m$  of sodium chloride at room temperature. The concentration  $m=2$  means 12 weight per cent salt in the water.



The concentration  $m$  of the salt may be different in separate water phases even in thermodynamical equilibrium. The pore-water pressure  $p_\ell$  will then have different values in different water phases in the pores.

Equations (3), (4), and (5) lose their meaning in extremely small liquid phases. Consider water in a pore with linear dimensions of say 15  $\text{\AA}$ , which corresponds roughly to the thickness of five layers of water molecules. The influence of the solid walls will give a more complicated state of tension than that of a simple isotropic pressure or tension. The description requires the use of a full tension tensor. The equations for thermodynamical equilibrium are valid for a point in the liquid water as long as a simple isotropic

state of tension, characterized by a hydraulic pressure or tension  $p_l$ , prevails.

The equilibrium conditions are valid locally also during a dynamical flow process for which the states through the material change with time. A liquid phase and a neighbouring gas phase lie so close together that the local deviations from equilibrium ought to be exceedingly small, even during the most violent dynamical process.

## MOISTURE FLOW

Water molecules may flow or diffuse in liquid phases, gas phases and along adsorbed water layers on the pore walls. We will in the following discussion disregard the possibility of migration of water in the adsorbed water layers on the pore walls.

The water of a liquid phase will move when there is a gradient in the local pore water pressure  $p_l$ . A gradient in the local gas pressure  $p_g$  will cause a convective flow of the gas with its water-vapour, while a gradient in the water-vapour pressure  $p$  will cause a diffusion of vapour through the gas phase. Moisture flows are caused by gradients in  $p_l$ ,  $p_g$  and  $p$ . These are (excluding the possibility of surface migration) the three main direct physical causes for moisture movements in the porous material.

Convective gas flows require gradients in the gas pressure  $p_g$ . The main cause, besides externally applied pressure differences over the material, for changes in  $p_g$  is temperature changes. A temperature change in a gas phase of essentially constant volume entails a minor pressure change in accordance with the gas law. The pressure changes when the temperature is raised are mainly due to evaporation of water so that the vapour pressure increases considerably. The vapour pressure will roughly change as the saturation vapour pressure  $p_s(T)$ , as long as the material contains enough liquid water. A rapid solar heating of a humid wall to say  $50^\circ\text{C}$  may raise the vapour pressure to  $p=0.25 \cdot 10^5$  Pa. This will give an increase of the gas pressure  $p_g$  with 25 per cent. An extreme heating of a wall of concrete during a fire to say  $200^\circ\text{C}$  may increase the gas pressure  $p_g$  in the pores from  $1.0 \cdot 10^5$  Pa up to  $16 \cdot 10^5$  Pa. The intensity of the convective gas flows through the material depends on the degree of free gas passages through the pore system. An increasing extension of the liquid phases will diminish the gas passages and lower the gas flow. In conclusion we have that a high moisture flow intensity in the form of convective gas flow requires considerable and rapid temperature changes (or externally applied pressures).

We will now limit the discussion to the case when gas pressure gradients and convective gas flows are negligible. There are water-vapour flows due to gradients in  $p$  and liquid flows due to gradients in  $p_l$ . The flow pattern on the pore level may be extremely complicated. There is vapour diffusion in gas phases and convective flows through liquid phases. Water evaporates from and condensates on myriads of water surfaces in the pores. These flows in the phases and between phases affect each other. The heat of evaporation and condensation, the energy flows, and in particular the heat flows also affect the moisture flow pattern.

A particular water molecule may diffuse through a gas phase, condensate on a liquid surface, flow through the liquid phase, evaporate into a new gas phase, and so on. A separation of the moisture flow in a vapour and a liquid component is not a very precise concept. It is only the total moisture flow that has a precise measurable meaning.

The state in a point of the material is given by two independent variables, when we assume constant gas pressure  $p_g$  and neglect effects of salts. A natural choice, from a physical point of view, of independent variables is  $p$  and  $p_l$ .

Let  $F_{wl}$  and  $F_{wv}$  denote the liquid and vapour flows respectively. The total moisture flow  $F_w$  is equal to  $F_{wl} + F_{wv}$ . This splitting should be used with caution since we cannot measure these two flows separately. The vapour and liquid carry their respective internal energies  $e_v$  and  $e_l$  (J/kg). The considerable difference  $e_v - e_l$  is equal to the latent heat of evaporation (at constant volume). This gives a convective contribution  $e_v F_{wv} + e_l F_{wl}$  to the total energy flow  $F_e$ . There are also contributions from pressure work in the displacement of the masses. Finally there is a heat flow component  $F_q$ . Neglecting contributions from pressure work we have

$$F_e = F_q + e_v F_{wv} + e_l F_{wl} \quad (7)$$

This equation may be regarded as a definition of what we call the heat flow. The physical content of the formalism lies in the expression for  $F_q$ ; for example when we postulate Fourier's law:

$$F_q = -\lambda \frac{\partial T}{\partial x} \quad (8)$$

Here  $\lambda$  is the heat conductivity. It will depend on the moisture content  $w$ .

The trouble with (7) is that we do not know the separate flows  $F_{wv}$  and  $F_{wl}$ . A more rigorous approach is to postulate directly for  $F_e$  a linear expression in gradients of state variables in the same way as we did for the moisture flow  $F_w$ . The energy transport coefficients in this linear expression should in principle be measured directly. This however poses great experimental difficulties.

Fortunately, the moisture flows in porous building materials are in most applications very small. The contribution to the energy flow from the moisture flow is then negligible compared to the ordinary heat flow. The energy flow  $F_e$  is with good accuracy equal to the heat flow  $F_q$ . The change in the energy content due to changes in the moisture content is then also negligible. The energy conservation equation becomes the usual heat conduction equation:

$$\frac{\partial e}{\partial t} = C \frac{\partial T}{\partial t} \quad F_e = F_q = -\lambda \frac{\partial T}{\partial x} \quad C \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( \lambda \frac{\partial T}{\partial x} \right) \quad (9)$$

Here  $C$  is the heat capacity (at constant moisture content) per unit volume of porous material (J/m<sup>3</sup> °C). The coefficients  $C$  and  $\lambda$  will depend on the moisture content  $w$ .

Let us now briefly consider the general case when the influence of the moisture flow on the energy balance cannot be neglected. The difficulty stems from the fact that we do not know the separate vapour and liquid flows. We may alternatively say that the difficulty is due to the fact that we do not know the net rates of evaporation or condensation in each point of the material. The heat of evaporation affects the energy equation in an uncontrollable way.

It should be noted that it is physically unsound and arbitrary to make direct assumptions about the distribution of heat generation and consumption due to condensation and evaporation within the material. The gradients in state variables, which arise during the dynamical process, induce certain mass flows in accordance with the transfer coefficients. The evaporation and condensation is then governed by the requirement that local equilibrium shall prevail. For example, an excess net inflow of water vapour to a small region of the material will be accompanied by an appropriate rate of net condensation so that



local thermodynamical equilibrium is maintained.

## HYSTERESIS

The relation between moisture content  $w$  and the state of the moisture, given for example by  $p/p_s$  or  $p_g$ , often exhibit pronounced hysteresis for building materials. The amount and distribution of the pore water in the pores depend not only on the actual state of the moisture but also on the previous moisture history. We get a new branch for the relation each time the direction of the change in state of the moisture is altered. The moisture content  $w$  depends on the state of the moisture and on all preceding turning points or states at which the direction of change was altered. Moisture transport coefficients may also exhibit this complicated type of dependence.

Special complications arise when the moisture flow is related to the gradient of the moisture content  $w$ . Let  $D_w$  denote the corresponding transport coefficient. The problems are due to the fact that moisture flows are caused by gradients of intensive state variables like  $p$  and  $p_g$ , and not by gradients of  $w$ . The dependence of  $D_w$  on the previous moisture history will become very intricate because of the moisture hysteresis. Consider as an illustration an equilibrium situation where the moisture content  $w$  varies through the material due to hysteresis. There are not any flows, and the intensive state variables are constant through the material. The transport coefficient  $D_w$  must in this completely ordinary situation be zero, since the gradient of  $w$  is different from zero. The description with  $D_w$  as transport coefficient should only be used in monotonous moisture processes, where there is a single relation between the moisture content and the state of the moisture.

These hysteresis effects are caused by irreversible processes when the moisture content changes locally in a pore. Consider a situation when a part of a pore is filled by pore water in an irreversible process. This part of the pore cannot be emptied by retracing the process in the opposite direction, because of the irreversible character of the process. The pore has to be emptied in some other way during other conditions. The pore water can therefore be distributed in different ways in the pores depending on the previous moisture history.

There are many possible types of irreversible processes. A water surface in a pore may encounter an obstacle. An example is when two water surfaces meet and coalesce irreversibly to a single water surface in a new position. Different types of thermodynamical instabilities also cause irreversibilities. The water surfaces in the pores may reach an unstable position. Then the surface moves swiftly to a new, stable position.

The various branches for the relation between the moisture content and the state of the moisture, as well as the corresponding different branches for moisture transport coefficients, have to be known when a dynamical process is numerically simulated. The turning points, where the direction of the change in the state of the moisture is altered, must be saved for each point in the material in order to be used in the subsequent calculations. Hysteresis effects cause considerable complications to dynamical moisture and energy flow processes.

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