MASS TRANSFER IN NATURAL ORGANIC MEDIA

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Mass-transfer mechanisms are considered for organic rheological media at positive and negative temperatures. The transfer can be controlled in a powder by physicochemical modification.

Water transport in a granular material is a physicochemical process that includes various surface and internal phenomena, whose type is in turn governed by the state, the properties, the phase ratios, and the mass-transfer rates. Basic principles have been given [1-3] for the physics of water transport in peat systems, which are the most common organic materials. Here we consider only recent results on the mechanisms governing the transport of water and ions in peat, peaty soils, and subsoils, including methods of influencing those processes.

The wetting-film thicknesses and total bound-water contents are [1, 4] dependent on the surface charges, which are very sensitive to pH in the dispersion medium. As the pH increases and correspondingly the charge, the bound-water content in a peat system rises [4], i.e., the pH affects the relation between the types of water, and that itself influences the mobility. The diffusion coefficient a_w for water in peat at $2 \lesssim \text{pH} \lesssim 9.5$ varies by about two orders of magnitude [4], and as the pH falls, a_w rises, whereas the thermal mobility δ falls. Conversely, a_w falls and δ increases as the pH rises [4].

Isothermal water transport in a connected-pore system in general represents the flow of the dispersion medium as capillary or film water in the gaps and over the surfaces in response to a potential gradient. If water molecules or solutes are to move within the bulk, they have to overcome interactions with their neighbors, and also surface forces move in the bound-water layers. Naturally, the mobility of pore water within the surface-force range is much less than in the bulk. The bound-water content in a peat system may be reduced because the surface of the solid goes over to the isoelectric state as the pH is reduced, which increases the mobility and the transport of ions and water-soluble compounds in the water to the mass sink, i.e., to the phase-transition (evaporation) zone [4].

As the pH rises, the water diffusion, conductivity, and water-soluble compound migration fall in a peat system [4], but there are also effects on the water and solutes from changes in structure and ion-exchange capacity, since the organic components of the peat swell as the pH rises, which reduces the active capillary network and thus the water conductivity. The converse occurs when the pH is reduced. The friable humus formations in the peat prevent compact coagulation and activate the capillary network, and thus also the water transport. The variations in a_w [4] in peat systems at $\text{pH} \leq 4$ may [3] be described as colloidal connected-pore type, while those occurring at $\text{pH} > 4$ are typical of colloidal materials. Also, at low pH, the concentrations of the ions in the dispersion medium increase, whereas they are reduced at high pH because ions are transferred from the exchangable state to the solution or from the solution to that state. That process in the first case accelerates the redistribution and water migrates [4, 5], and, in the second, reduces it.

The water transport in a natural dispersed material such as peat is usually characterized by nonisothermal conditions, and thermal water transport occurs in a connected-pore system in the three-phase state [3], which corresponds to the highest thermal mobility due to temperature gradients. At low water contents ($U \to 0$), the mobility is reduced because the binding energy to the solid increases, and in the two-phase state, one can get only thermal mass circulation without redistribution in the bulk ($dU/dT|_{i=0} = 0$). Also, as U increases, the liquid–gas interface area is reduced, and that governs the thermal water transport under surface tension gradients. The latter produces a flow of water on account

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of the gradient in the capillary potential. Then the highest thermal mobility in the medium ($dU/dT|_{i=0} = \delta$ [3]) corresponds to a state where the pores are not completely filled with water and the liquid–gas interface is fairly extensive [6]. The water in the pore space is then split up by short film areas, and the thermal-gradient coefficient δ is dependent on the thermal mobility there [4].

The boundary-layer properties thus have marked effects on the thermal transport rate in a connected-pore body, in addition to effects from the phase ratio [1-3, 6, 7].

There is [7] a formula relating the maximum thermal-gradient coefficient δ_{max} for a peat system to the pH:

$$\delta_{\max i}|_{\rho} = \delta_{\max 0} + K(\mathrm{pH}_i - \mathrm{pH}_0). \tag{1}$$

As the pH increases, the water-content range giving vigorous thermal transport expands, and the peak δ shift to higher water contents [7]. That range expansion, with similar rates of thermal transport within the range, is virtually impossible to provide in peat by physical methods such as altering the phase ratio or the structure. For example, for $U > 1.6$ kg/kg, δ in a peaty soil with pH 7.4 exceeds that in an analogous soil at pH 4.7 by about two orders of magnitude [4]. The same applies to materials whose structure is independent of pH (quartz sand) [6], i.e., the changes in thermal mobility in a granular material are to some extent related to the thicknesses of the wetting layers on the particles with varying charge densities. As a result, the bound water thermal flux increases with pH and there is increased redistribution in a peat system [6, 7].

The water is not only the solvent but also the medium that transports the solute. The transport mechanism and the mass-transfer parameters govern the migration rates and redistribution for the solutes, and particular interest attaches to electrolyte migration in thin surface films, while ion migration provides a method of obtaining information on the boundary-layer structures and the hydrodynamics under various transfer conditions.

Water transport in a natural open-pore medium such as peat can be divided into two aspects: transport of the solvent (water) and solute migration, which are closely connected, with the water transport occurring in liquid or vapor phases, but the solutes only in the liquid. With three-phase states, the transport in liquid form represents the migration of thin layers over the components. In that mechanism, the solute migration is influenced by the features of the water boundary layers and also by the water content bound by surface forces.

The ion redistribution in peat increases as the pH falls [6]. Near the neutral point, the main ion-flow direction coincides with the heat-flux direction in a peat system. In the acid range, the salt migration is directed to the water phase-transition zone. Also, in nonisothermal transfer, the ion migration in a peat occurs in the wet and hygroscopic states and ceases at $U \leq 0.25U_{mh}$ (maximally hygroscopic state) [4].

Thermal transport in a peat is [1-3, 7] due in the main to three components of the thermal flux: vapor thermal diffusion, bound-water thermal transport, and thermocapillary flow. The general direction of the thermal flux is that of the heat flux and, in turn, the thermal water transport tends to counteract the diffusion flux and the film transport produced by the gradient in the wetting pressure, as those factors tend to equalize the distribution in the peat and to bring it to a state of diffusion equilibrium.

In a closed system, the water circulates spontaneously [4, 7]: thermal diffusion in the vapor TDV − bound-water thermal flux BMF + thermocapillary flux TCF $\gtrless$ diffusion flux DF + film flow FF under wetting pressure gradients. The diffusion is most rapid in the medium least bound by the surface forces, where a_w is maximal, i.e., in water layers directly at the liquid–gas boundaries. In the stationary state, when the total flow in unit volume is zero, the thermal flux in the wetting films will be vigorous not over the entire profile but only in the layers where the BWF is greater than the TCF. The film thickness controls whether the BWF occurs in the layers directly at the liquid–gas boundaries (thin films) or within the wetting films (thick films, capillary water). The water loss from the material is higher the greater the BWF [4].

At low pH (low bound-water content), the BWF contribution to the thermal flux is reduced, but the DF and FF increase. The TDV becomes decisive. This involves rapid ion transport into the phase-transition zone [4], i.e., change in the direction of preferential ion transport in the material.

The wetting films are highly mobile at the liquid–gas boundaries, where the negative charges on the organic particles result in maximum anion density and, therefore, the anions are active in migration. The anion migration in water transport in peat is usually much greater than that of the cations: $NO_3^- \gg NH_4^+ > Na^+ > K^+ > Ca^{2+}$ [4, 5], and the differences between anion and cation migration rates are most prominent in isothermal transport, where the water flux is uniform in direction.

Water-soluble organic compounds are important components in pore solutions in peat systems, which are mainly carbohydrates and humic substances. Those compounds migrate only in the DF and FF directions and are not transported by the bound-water thermal flux [4], so those compounds are localized in the medium not bound by surface forces, i.e., in the pore water (moist state) or in the wetting films at the interfaces (hygroscopic state).

The equilibrium is also determined by ion exchange, adsorption, and electrokinetic phenomena, which occur within the interphase layers and affect the properties and the transport rate. The pore-solution compositions affect [4] the ion redistribution in the thermal transport in a peat system. The cation activity sequence is $NH_4^+ > Li^+ > K^+ > Na^+$, and the anion one $Cl^- > I^- > Br^-$. The mechanism for cations at a given ionic strength involves the selectivity of the ion exchange, while for the anions the mechanism is affected by the diffusion mobility of the water [8].

Water can thus show high thermal mobility in a porous material when the wetting films are thick (much bound water), which requires elevated surface potentials identical in sign between the solid and the gas [9]. In turn, the water diffusion mobility increases when the boundary layers are thinner, i.e., when there is less bound water and the interfaces go over to the isoelectric state [4].

One can control water and ion transport in peat by means of surfactants SA because they are adsorbed selectively at boundaries. Anionic surfactants (ASA) reduce a_w [4, 8], while cationic ones (CSA) increase it. At the same time, ASA increase the thermal mobility of the water, while CSA reduce it in the concentration range where the surface charge is unaffected. That is a regular SA mechanism because ASA in peat raise the negative charges and correspondingly the bound-water contents [4], while the positively charged CSA reduce the negative potential and the bound water. Correspondingly, the ion migration is affected: ASA will reduce the redistribution during water transfer, while CSA accelerate it [4].

Water-soluble high-molecular compounds (HMC) are highly effective in influencing boundary layers and transfer in peat [8], since they not only affect the surface charges but also raise the water viscosity and the bound-water content [7]. Anionic HMC reduce the mobility very considerably and thus result in less ion migration in peat [8]. Cationic HMC have effects similar to CSA at low concentrations. At higher HMC concentrations, they increase the water diffusion and the ion migration rate to a maximum, which corresponds to the isoelectric state (minimum bound water), and then cause a fall because of the charge reversal [5, 8]. Nonionic HMC have much less effect than ionic ones.

The mass transport when a natural organic system freezes is qualitatively different from nonisothermal mass transport at positive temperatures.

In a peaty soil, the water flux from the thawed zone into the frozen one is proportional to the temperature gradient [10, 11], and the coefficient of proportionality has been called the frozen-zone thermal water permeability coefficient K, so if this is known along with the temperature gradient in the frozen zone, one can determine the water flux to the zone boundary:

$$q_w = -K_f \frac{\partial T}{\partial x} . \tag{2}$$

Theory shows [12, 13] that the main driving forces for water transport in the frozen material are temperature and hydrostatic-pressure gradients:

$$q_w = \alpha_{11}\rho_1 \left(\Delta P + \rho_2 L \frac{\Delta T}{T}\right) . \tag{3}$$

Then (3) enables one to determine the equivalent relation between the hydrostatic-pressure and temperature gradients that can be used to correct for the effects of hydrostatic pressure on the mass transfer on freezing.

If there is cross-linking, hydrostatic-pressure gradients may arise in the frozen zone, which on the one hand will tend to break the links to provide for excess ice deposition and on the other will reduce the water migration through the unfrozen layers. We assume that the ice in the frozen zone accumulates at a temperature T_i, at which the hydrostatic pressure due to the temperature gradient exceeds the local strength P_s in the adhesion, and on the basis of the equivalent relation between the gradients in (3), we get a formula for the water migration flux from the thawed zone to the frozen one:

$$q_w = -\left[\frac{(T_\xi - T_i) - (P_s - P_0)\dfrac{T_0}{\rho_2 L}}{\displaystyle\int_{T_\xi}^{T_i} \frac{1}{a_w(U_\ell)\rho_{sk}\dfrac{dU_\ell}{dT}}\,dT}\right]\frac{dT}{\partial x}. \qquad (4)$$

From (4), the phenomenological coefficient k_f for the zone boundary is

$$K_f = -\left[\frac{(T_\xi - T_i) - (P_s - P_0)\dfrac{T_0}{\rho_2 L}}{\displaystyle\int_{T_\xi}^{T_i} \frac{1}{a_w(U_\ell)\rho_{sk}\dfrac{dU_\ell}{dT}}\,dT}\right]. \qquad (5)$$

The flux from the thawed zone to the frozen one is thus determined not only by the diffusion coefficient for the unfrozen water but also by the strength characteristics. This is one of the simplest forms of model that can be realized in the freezing of a natural dispersed material. In the more general case, one can consider the migration mechanisms on freezing only by incorporating complicated rheological processes in the frozen zone produced by the pressure gradients. However, no matter what the transport mechanisms, K_f can be used in the absence of external loads to calculate the water redistribution on freezing.

We now consider a homogeneous semiinfinite medium consisting of a moist soil at an initial temperature $T_0 > T_f$ with water content U_0. At the start, a constant temperature $T_c < T_f$ is established at the surface of the soil. The freezing is accompanied by water migrating from the thawed zone to the frozen one and by swelling. The heat and mass transfer characteristics are given as fucntions of the water content and temperature. One has to derive the temperature and water-content distributions as functions of time and to determine the swelling. To simplify the treatment, we assume that the ice segregation occurs at the freezing front and that the swelling is determined by the increase in the ice content of the frozen zone due to water migration.

We introduce a coordinate system in the frozen zone linked to the mobile soil surface and, in the thawed one, a system linked to the initial position of the surface. We denote these coordinates correspondingly by x and y. One transfers from one system to the other via

$$y = h(\tau) + x, \qquad (6)$$

in which $h(\tau)$ is the swelling.

We assume that the swelling is proportional to the advance of the freezing zone into the thawed zone ξ_x and introduce the relative swelling coefficient

$$E_s = h(\tau)/\xi_x(\tau). \qquad (7)$$

There is a functional relation between the frozen zone relative to the mobile surface ξ_y and the advance of the freezing front ξ_x:

$$\xi_y = (1 + E_s)\,\xi_x. \qquad (8)$$

The mathematical formulation is

$$\frac{\partial T_1}{\partial \tau} C_1(U_1) = \lambda_1(U_1)\frac{\partial^2 T}{\partial y^2}, \quad 0 < y < \xi_y(\tau); \qquad (9)$$

$$\frac{\partial T_2}{\partial \tau} C_2(U_2) = \lambda_2(\bar{U}_2) \frac{\partial^2 T}{\partial x^2}, \ \xi_x < x < \infty; \tag{10}$$

$$\lambda_1(U_1) \left.\frac{\partial T_1}{\partial y}\right|_{\xi_y(\tau)} - \lambda_2(\bar{U}_2) \left.\frac{\partial T_2}{\partial x}\right|_{\xi_x(\tau)} = Q_\mathrm{p}(U_1, \rho^\mathrm{f}_\mathrm{sk}) \frac{d\xi_y}{d\tau}; \tag{11}$$

$$T(x, 0) = T_\mathrm{c}, \ T(0, \tau) = T_\mathrm{c}, \ T_1|_{\xi_y(\tau)} = T_2|_{\xi_x(\tau)} = T_\mathrm{f}, \ y = h(\tau) + x; \tag{12}$$

$$Q_\mathrm{p}(U_1, \rho^\mathrm{f}_\mathrm{sk}) = L\rho^\mathrm{f}_\mathrm{sk} U_\mathrm{i}(T_\mathrm{f}); \ U_\mathrm{i}(T) = U_1 - U_\ell(T); \tag{13}$$

$$\frac{\partial U_2}{\partial \tau} = a_w \frac{\partial^2 U_2}{\partial x^2}, \ \xi_x(\tau) < x < \infty; \tag{14}$$

$$q_w|_{\xi_x(\tau)} = -K_\mathrm{f} \left.\frac{\partial T_1}{\partial y}\right|_{\xi_y(\tau)}; \tag{15}$$

$$\left.\frac{\partial U_2}{\partial x}\right|_{\xi_x(\tau)} = \left(K_\mathrm{f} \left.\frac{\partial T}{\partial y}\right|_{\xi_y(\tau)}\right) \Big/ a_w \rho_\mathrm{sk}, \tag{16}$$

$$U(x, 0) = U_0.$$

If we know how the thermophysical characteristics and phase-transition heat are dependent on the water content and skeleton density, and if the swelling is known, the solution for the temperature pattern is

$$T_1(y, \tau) = T_\mathrm{c} + \frac{(T_\mathrm{f} - T_\mathrm{c}) \operatorname{erf}(y/2\sqrt{a_1(U_1)\tau})}{\operatorname{erf}[\beta_y/2\sqrt{a_1(U_1)}]}, \tag{17}$$

$$T_2(y, \tau) = T_0 - \frac{(T_0 - T_\mathrm{f}) \operatorname{erfc}(x/2\sqrt{a_2(\bar{U}_2)\tau})}{\operatorname{erfc}[\beta_x/2\sqrt{a_2(U_2)}]}, \tag{18}$$

$$\frac{\lambda_1(U_1)(T_\mathrm{f} - T_\mathrm{c})}{\sqrt{a_1(U_1)} \operatorname{erf}[\beta_y/2\sqrt{a_1(U_1)}]} \exp\left(-\frac{\beta_y^2}{4a_1(U_1)}\right) - \\ - \frac{\lambda_2(\bar{U}_2)(T_0 - T_\mathrm{f})}{\sqrt{a_2(\bar{U}_2)} \operatorname{erfc}[\beta_x/2\sqrt{a_2(U_2)}]} \exp\left(-\frac{\beta_x^2}{4a_2(\bar{U}_2)}\right) = \frac{Q_\mathrm{p}(U_1, \rho^\mathrm{f}_\mathrm{sk})\sqrt{\pi}}{2} \beta_y, \tag{19}$$

$$\beta_y = (1 + E_\mathrm{s}) \beta_x, \tag{20}$$

in which β_y and β_x are parameters defining the advance of the freezing front in accordance with $\xi_y = \beta_y\sqrt{\tau}$ and $\xi_x = \beta_x\sqrt{\tau}$.

To determine the thermophysical characteristics in the frozen zone, which are dependent on the total water content there, one has to consider the water redistribution on freezing. This gives formulas for the water distribution in the thawed zone and the ice accumulation in the frozen one:

$$U_2 = U_0 - \frac{\sqrt{\pi}\alpha K_\mathrm{f}}{\rho^\mathrm{t}_\mathrm{sk}\sqrt{a_w}} \exp\left(\frac{\beta_x^2}{4a_w}\right) \operatorname{erfc}\left(\frac{x}{2\sqrt{a_w\tau}}\right), \tag{21}$$

$$\Delta U_1 = \frac{2\alpha K_\mathrm{f}}{\rho^\mathrm{t}_\mathrm{sk}\beta_x} - \frac{\sqrt{\pi}\alpha K_\mathrm{f}}{\rho^\mathrm{t}_\mathrm{sk}\sqrt{a_w}} \exp\left(\frac{\beta_x^2}{4a_w}\right) \operatorname{erfc}\left(\frac{\beta_x}{2\sqrt{a_w}}\right). \tag{22}$$

Parameter α characterizes the temperature gradient in the frozen zone at the freezing front and is defined by the conduction equation

$$\alpha = \left.\frac{\partial T}{\partial y}\right|_{\xi_y} \Big/ \sqrt{\tau}. \tag{23}$$

If the solution to the conduction equation is self-similar, the water content in the thawed zone at the freezing front and the water content in the frozen zone will be constant, i.e., the mass-transfer solution will also be self-similar and the water redistribution on freezing does not disrupt the self-similar feature in the temperature pattern.

If we know the increase in water content in the frozen zone and assume that the swelling is due to the volume of the water migrating into the frozen zone and becoming ice and also due to the volume change in the initial amount of water on the formation of ice, we have

$$E_s = \frac{\Delta U \rho^{t}_{sk}}{\rho_2} + E_0, \tag{24}$$

in which E_0 is the swelling coefficient due to the change in the volume of water on conversion to ice without allowance for the migration.

In general, one can calculate the relative swelling coefficient only from additional information on the structure and texture of the frozen soil.

System (17)-(24) is coupled and gives a solution for the heat and mass transfer on freezing in wet finitely-divided materials accompanied by swelling. One can solve the system by successive approximation if one calculates the behavior of the temperature pattern while neglecting the migration and swelling and then incorporates the changes in thermophysical characteristics and phase-transition heat due to the migration and swelling and then repeats the calculation on the temperature pattern. Practical calculations on peat show that the entire operation may be repeated 2-4 times to obtain consistent values of β_y, ε_x, ΔU, and E_s.

These results give a sound basis for incorporating the mass transport mechanisms in such materials for the purposes of analysis, simulation, and calculation on analogous phenomena in natural media.

NOTATION

a_w, diffusion coefficient in thawed soil; δ, thermal-gradient coefficient; K, coefficient characterizing the sensitivity of the thermal water flux to the acidity of the dispersion medium; q_w, water flux to frozen zone; K_f, thermal water conductivity coefficient for frozen soil zone; L, latent heat of phase transition from water to ice; ρ_1, density of water; ρ_2, density of ice; ∇P and ∇T, hydrostatic-pressure and temperature gradients; α_{11}, coefficient characterizing the hydrodynamic conductivity from unfrozen water layers; P_0, hydrostatic pressure at boundary between thawed and frozen zones; h, swelling; T_1 and T_2, temperatures in the frozen and thawed zones; T_c, temperature at soil surface; T_0, initial temperature; T_f, freezing point of water in soil; τ, time; $C_1(U_1)$ and $\lambda_1(U_1)$, bulk specific heat and thermal conductivity of thawed soil; $C_2(U_2)$ and $\lambda_2(U_2)$, bulk specific heat and conductivity of thawed soil; $a_1(U_1)$ and $a_2(U_2)$ thermal diffusivities of frozen and thawed zones in soil; $Q_p(U_1, \rho_{sk}{}^f)$ bulk heat of phase transition on frozen soil; U_1, total water content in frozen soil zone; U_i, ice content of soil; $U_\ell(T)$, amount of liquid water; U_2, water content in thawed soil zone; U_2, average water content in thawed zone; U_0, initial water content in soil; $\rho_{sk}{}^t$ and $\rho_{sk}{}^f$, skeleton densities for thawed and frozen soil.

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KINETICS AND MECHANISM OF SOLIDIFICATION AND HEAT TRANSFER OF EPOXY COMPOSITES

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A model is constructed of the kinetics of epoxy resin solidification of isomethyl tetraphthalate anhydride in the presence of a ternary amine. Parameters of the kinetic equation are determined from tests and a computation is performed for an epoxy resin with solidifier of amine type. The method of space—time separation permitted estimation of the influence of rheokinetics on the flow and heat transfer of the partially polymerized fluid.

Plastics are replacing metal, wood, cotton, wool, silk, etc. more and more at present, especially in thermal and electrical engineering, electronics, construction and transport, medicine and agriculture. Until recently, primarily thermal plastics synthesizable from simple organic compounds, monomers, were used. Prior to molding the thermal plastics were transformed into the flowing state for reworking into articles. Reaction plastics, obtained from oligomers of resin type with a small degree of polymerization, are slightly viscous in the initial state and expenditures on their reworking are not large while the molecular mass of the end product is practically infinite. Joining of oligomer complexes is initiated by special admixtures (solidifiers). The appearance of rapidly solidifying oligomer composites set up the beginning of a new progressive industrial technology. Obtaining both the material and the article within a very short time is combined in a single cycle during chemical molding.

Epoxy resins (ER) are evolved efficiently with good mechanical, electrical, and adhesive properties as well as insignificant shrinkage during solidification. They are applied diversely in the production of composite materials, coatings, glues, etc.

A solid substance with developed cross bonds is formed from a liquid mixture of monomers and oligomers during solidification (polymerization, polycondensation). Much heat is liberated here. The chemical reaction rates depend radically on temperature; therefore, the heat transfer governs the course of the solidification process and the quality of the end product to a decisive extent.

The spatial homogeneity of the viscosity and degree of transformation of the composite is of exceptional importance during pressure-casting of a partially solidified polymer composite. The kinetic model of the solidification and the spatial evolution of the temperature fields must be known to control the technological processes.

A kinetic model can be determined by two methods. If the chemical solidification mechanism and the change in enthalpy of the individual reactions are known, then the reagent concentration and heat liberation can be expressed by a system of differential equations. It reduces, often enough, to one kinetic equation connecting the reaction rate (the rate of

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