



ON SOME ASPECTS OF A NONLINEAR CONVECTION-DIFFUSION EQUATION ARISING IN HEAT-INDUCED MOISTURE TRANSPORT IN POROUS MEDIA

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Abstract—This paper focuses on the problem of the heat-induced moisture movement in a porous medium. In particular, the paper addresses the nonlinear convection–diffusion processes arising from such moisture transport under steady thermal gradients. The character of the governing differential equation is examined in relation to its solution structure. Conclusions with regard to the residual moisture distribution within an annular porous medium are established.

1. INTRODUCTION

The topic of heat-induced moisture transport in porous media has been studied quite extensively in connection with potential applications to chemical engineering and agricultural engineering. In recent years, however, there has been a renewed interest in this topic particularly in relation to extraction of geothermal energy resources, heat exchangers which are installed in geological media and in the disposal of heat generating nuclear fuel wastes in geologic media. The motivation for this research stems from the potential application of the results to the geologic disposal of heat emitting nuclear fuel wastes in a geologic medium. In such an endeavour, canisters containing stacked heat emitting nuclear fuel bundles will be emplaced in disposal galleries created in deep vaults mined into stable geological formations. The waste canisters will be housed in boreholes which are drilled into the base of a vault system. In order to isolate the heat emitting waste containers from the host rock an engineered geological barrier will be employed. An example of such an engineered geological barrier is a moist mixture of bentonitic clay and crushed sand which will be compacted within an emplacement borehole (Fig. 1). The heat emitting waste container will be placed in cavities drilled into the compacted geological buffer. The pliable, relatively impermeable buffer material is expected to isolate the waste container from water migration from the rock and suppress the detrimental effects of the corrosive ground water. In addition, the buffer should isolate the waste container from the rock mass. In doing so the buffer must maintain its integrity without development of shrinkage cracks which can result from excessive heat-induced moisture movement. This aspect is crucial to the waste management effort since the occurrence of cracks and fissures within the buffer will render radionuclide transport via flow processes rather than via diffusive mass transport with buffering action provided by an intact bentonitic clay mass. The time scales involved in the two processes are such that a cracked buffer which can result from heat-induced moisture transport will render the waste disposal system ineffective, particularly in the event of a premature damage of a waste container the rate of release harmful radionuclides through a cracked buffer is expected to be detrimental to the environment.

In this paper we focus on the processes that are associated with heat-induced moisture movement in a nondeforming porous geological material. In particular attention is restricted to the study of such processes which can occur within an annular region in a radially symmetric

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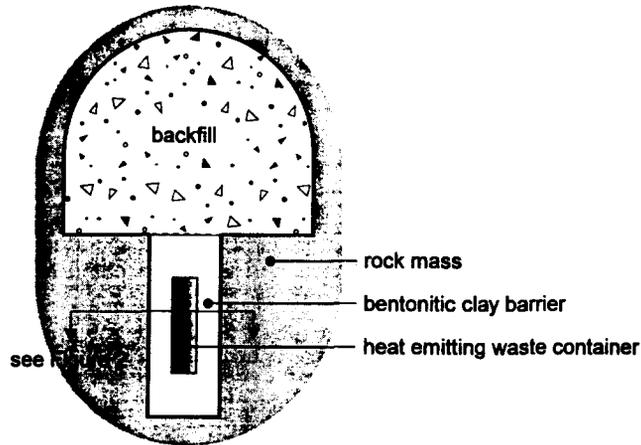


Fig. 1. Schematic view of a waste emplacement borehole.

fashion (Fig. 2). The radially symmetric condition is assumed to be representative of the hydro-thermal phenomena which can be present in the annular space of porous geological buffer region between the heat emitting waste container and the host rock mass. The objective of the study is to provide some general guidelines which can establish the pattern of moisture redistribution within a porous medium which can result from a sustained heating process. In particular, the assessment of the moisture depletion effects is also prompted by observations on experiments involving moisture redistribution within one-dimensional buffer columns which are subjected to end-heating [1] and experimental simulations of the borehole emplacement configuration [2,3]. For example, the experimental results derived from one-dimensional column experiments indicate the following trends:

- (1) The time-dependent variation of the temperature distribution exhibits a negative temperature gradient. In particular, at the initial stages of heating, the absolute value of the temperature gradient is generally higher than at the steady-state condition.
- (2) At the the initial stages of operation of a disposal vault the moisture influx from the rock-mass to the buffer region will be largely restricted. Consequently the heating of the porous buffer region will result in the redistribution of the existing moisture as a result of the temperature gradient. The assessment of the stable moisture redistribution pattern is of particular interest to establishing the extent of drying within the porous medium. In particular, one-dimensional heater experiments [1] indicate that due to the heating the volumetric moisture content $\theta(r, t)$ increases monotonically to a local maximum and

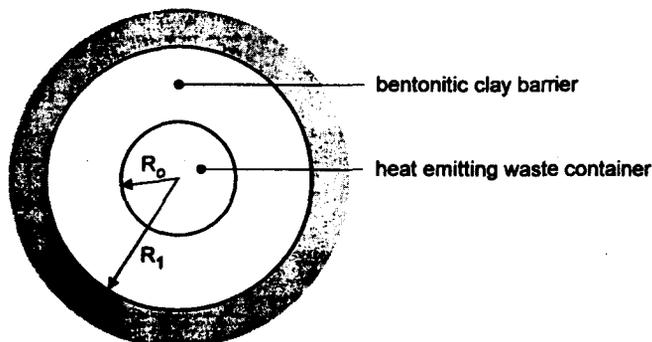


Fig. 2. Internal heating of annular clay barrier.

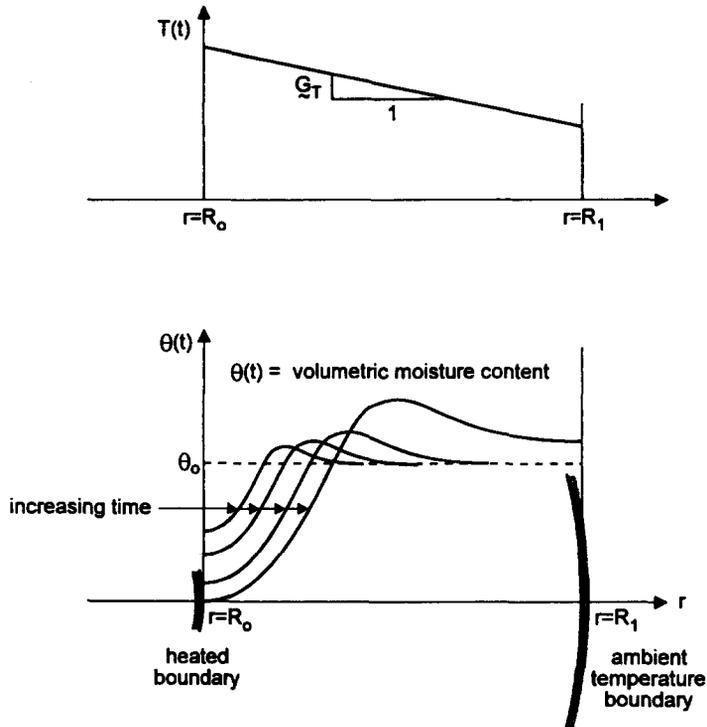


Fig. 3. Temperature gradient across the annular buffer region and the resulting moisture distribution pattern.

decreases monotonically to a stable initial value (Fig. 3). This observation is in contrast to the 'maximum principle' usually associated with purely diffusive phenomena.

The objectives of the research are to illustrate the following:

- The equation governing the heat-induced moisture movement within the porous medium can be described by a nonlinear convection diffusion type equation which yields a new class of solutions.
- The thermal gradients will cause the accumulation of moisture at an intermediate location within the buffer at the initial stages of heating; this process is however dependent on the geometry of the porous region.
- From the point of view of thermodynamics, the problem of moisture movement within the sealed porous medium is governed by the driving force which is expressed in terms of the temperature gradient and is supplemented by an additional relationship resulting from the second law of thermodynamics. This relationship provides the additional boundary condition which is unknown at the initial stage and is located at an internal surface within the buffer.

2. THE EVOLUTION OF THE TEMPERATURE FIELD

The process of heat transfer within the porous medium is assumed to be governed by the classical heat conduction equation. Experimental evidence suggests that the heat transfer process is relatively uninfluenced by the moisture redistribution process. In these experiments it is observed that the moisture transport due to heating induces shrinkage effects which in turn results in the development of zones of contact resistance particularly at the locations which are subjected to heating. These thermal jumps are however relatively small (these do not exceed 7% of the peak value of the temperature). Under the classical assumptions of heat transfer

which account for radiation conditions at infinity, the fundamental solution governing the heat transfer is given by (see, e.g. Carslaw and Jaeger [6], Smoller [7])

$$G(\mathbf{x}, t; \mathbf{y}, \tau) = \frac{1}{\sqrt{4\pi(t-\tau)^m}} \exp\left\{-\frac{|\mathbf{x}-\mathbf{y}|^2}{4\kappa(t-\tau)}\right\}, \quad t > \tau \quad (1)$$

where κ is the thermal diffusivity: $\kappa = k/\rho c$, k is the thermal conductivity and ρc is the heat capacity. A straightforward differentiation shows that G satisfies the heat equation with respect to the variables \mathbf{x} and t . The variable \mathbf{y} denotes the location of a unit source at time τ . With the aid of this fundamental solution, the function

$$T(\mathbf{x}, t) = \frac{1}{\sqrt{4\pi t^m}} \int_{R^m} \exp\left\{-\frac{|\mathbf{x}-\mathbf{y}|^2}{4\kappa t}\right\} \omega(\mathbf{y}) d\mathbf{v}_y \quad (2)$$

defines a differentiable solution of the heat equation in $R^m \times [0, \infty)$ with initial values

$$T(\cdot, 0) = \omega. \quad (3)$$

It is of particular interest to note that even in a steady state of heat conduction, there is a spatial gradient in the temperature field. This spatial gradient of the temperature will provide the driving force for the moisture movement within the porous medium. The spatial temperature gradient, the diffusivity properties of the system and the geometry of the region will ultimately govern the pattern of moisture redistribution.

3. GENERAL BALANCE EQUATION IN CONTINUUM THERMODYNAMICS

Let ψ be a measure of a thermodynamic quantity defined in a body $B(t)$. In a three-dimensional continuum it is assumed that ψ , which is treated as set function, i.e.

$$\psi = \psi(P), \quad P \subset B \quad (4)$$

is absolutely continuous with respect to the Lebesgue volume measure. Under the additional assumption on disjoint sets, a representation for the function ψ in the form of a volume integral of a density function ψ_v is derived and has the following form:

$$\psi(B) = \int_B \psi_v d\mathbf{v}. \quad (5)$$

Relationship (5) holds if the material volume B does not intersect with a singular surface S . In general, however, the singular surface can be embedded in, and interact with, a three-dimensional continuum. The singular surface can be a material surface or simply a conceptual moving surface located in B and carrying special properties. A moving surface embedded in a three-dimensional continuum divides the continuum into two regions for which the surface is a common boundary. Since the material of the interface exhibits properties entirely different from the three-dimensional continuum, it is assumed that the surface can be considered as two-dimensional continuum. In this general case the relation (5) is extended to the following integral representation for ψ (see Moeckel [8] and Müller [9]):

$$\psi(B) = \int_{B^+ \cup B^-} \psi_v d\mathbf{v} + \int_S \psi_s da \quad (6)$$

where the functions $\psi_v = \psi_v(\xi^k, t)$ and $\psi_s = \psi_s(u^\alpha, t)$ are volume and surface densities of ψ , while “ v ” and “ a ” are volume and area measures, respectively, for sets in ϵ^3 . Differentiability of ψ_v in B^+ and B^- is required in (6) and $B \equiv B^+ \cup B^-$ by definition.

A justification for the decomposition (6) can be given by arguments derived from measure theory and the interested reader is referred to Halmos [10] for basic concepts of measure theory and to Gurtin and Williams [11], Truesdell [12] and Murdoch [13] for their application

to surfaces embedded in a three-dimensional continuum. The existence of the second integral in (6) is evidence of the occurrence of surface effects associated with a concentration of the quantity ψ on a singular surface S , which can occur, for instance, at a boundary between dry and moist materials.

In continuum thermodynamics a general balance law for a body $B(t)$ is given in the following form (Moeckel [8]):

$$\frac{d}{dt} \psi_t = -\Phi(\psi_t) + P(\psi_t) + S(\psi_t), \tag{7}$$

where ψ_t , Φ , P and S are time dependent measures of thermodynamic quantities defined in $B(t)$. The measure $\Phi(\psi_t)$ is the efflux of ψ_t through the boundary of the body $\partial B(t)$, $P(\psi_t)$ is the production of ψ_t in $B(t)$ and $S(\psi_t)$ is the source (supply) of ψ_t in $B(t)$. In general Φ and P are given as constitutive quantities defined for various materials while $S(\psi_t)$ represents an external action on $B(t)$. The balance statement (7) is not universal and therefore not unique, it is, however, a sufficient extension of classical thermodynamics and it has been proved to have broad applications in modern engineering and technology problems [14–17]. It is usually assumed that the following suppositions hold:

SUPPOSITION 1. The balance law (7) holds for every subbody B .

SUPPOSITION 2. For thermodynamic quantities Φ , P and S the decompositions similar to (6) hold for any arbitrary subbody b and an instant t in the motion:

$$P(\psi_t(B)) = \int_{B^+ \cup B^-} \psi P_v \, dv + \int_{S(t)} \psi P_s \, da \tag{8}$$

$$S(\psi_t(B)) = \int_{B^+ \cup B^-} \psi S_v \, dv + \int_{S(t)} \psi S_s \, da \tag{9}$$

$$\Phi(\psi_t(B)) = \int_{\partial B^+ \cup \partial B^-} \psi E_v \, da + \int_{\partial S(t)} \psi E_s \, dl. \tag{10}$$

The quantities ψP_v , ψP_s , ψS_v , ψS_s , ψE_v and ψE_s are called the production densities in the volume and on the singular surface, densities of supply, and the flux densities of ψ_t , respectively. It is clear that ψ_v , ψE_v , ψP_v and ψS_v are not defined on $S(t)$; however, if we denote any of these functions by ϕ , it is assumed that the following limits exist:

$$\phi^+ \equiv \lim_{\substack{\xi \rightarrow \mathbf{x} \in S(t) \\ \xi \in B^+(t)}} \phi(\xi^i, t) \quad \text{and} \quad \phi^- \equiv \lim_{\substack{\xi \rightarrow \mathbf{x} \in S(t) \\ \xi \in B^-(t)}} \phi(\xi^i, t). \tag{11}$$

Also if $\phi(\epsilon^i, t)$ is continuously differentiable in $B^+(t)$ and $B^-(t)$, it is assumed that the above limits hold for spatial and time derivatives of ϕ to any desired order. At the surface $S(t)$, it is assumed that the above limits hold for spatial and time derivatives of ϕ to any desired order. At the surface $S(t)$ the difference $(\phi^+ - \phi^-) \equiv [\phi]$ is the jump of ϕ . Summarizing the representations (6), (8), (9) and (10) we can write the general balance statement (7) in the following form:

$$\begin{aligned} \frac{d}{dt} \left[\int_{B(t)-S(t)} \psi_v \, dv + \int_{S(t)} \psi_s \, da \right] &= - \int_{\partial B^+ \cup \partial B^-} \psi E_v^k N^k \, da \\ &\quad - \int_{\partial S(t)} \psi E_s^\alpha n_\alpha \, dl + \int_{B(t)-S(t)} \psi P_v \, dv + \int_{S(t)} \psi P_s \, da \\ &\quad + \int_{B(t)-S(t)} \psi S_v \, dv + \int_{S(t)} \psi S_s \, da. \end{aligned} \tag{12}$$

The general balance law expressed by (12) is a key relation to the formulation of local balance equations of modern thermodynamics. The interested reader is referred to Moeckel [8] and Müller [9]. The most important examples for balance equations in mechanics and thermodynamics are the equations of mass, momentum, energy, angular momentum and entropy. In this study, attention is focused on the balance equation governing conservation of mass.

A thermodynamical quantity ψ is said to be conservative if its production densities ${}^{\psi}P_v$ and ${}^{\psi}P_s$ vanish, since in that case the amount of ψ within B can only change by a flux through ∂B or by a supply from the exterior of B . The mass fluxes are zero because ${}^{\psi}E_v^k$ and ${}^{\psi}E_s^\alpha$ denote fluxes across a material surface or line respectively, i.e. a surface or line that moves with the particles. The supplies of mass and charge are also zero because mass and charge cannot be created inside a material volume by an action from the exterior. Consequently, the general balance law (12) for mass takes the form

$$\frac{d}{dt} \left[\int_{B(t)-S(t)} \rho_v dv + \int_{S(t)} \rho_s da \right] = 0, \quad (13)$$

i.e. the total mass is invariant in time. It turns out that relation (13) plays an important role in the modelling of problems related to heat induced moisture transport in porous media and it will be exploited in the proof of a main theorem of the paper. Another useful relation for further consideration is the formula related to the differentiation of a volume integral of a smooth function. The following holds: following Truesdell [12], if a scalar- or tensor-valued function f is continuously differentiable on the set $\bigcup_{t \in \tau} \{t\} \times B(t)$ and is continuous on the set $\bigcup_{t \in \tau} \{t\} \times \overline{B(t)}$, then

$$\frac{d}{dt} \int_{B(t)} f dv = \int_{B(t)} \frac{\partial f}{\partial t} dv + \int_{\partial B(t)} f \mathbf{v} \cdot \mathbf{n} da \quad (14)$$

where \mathbf{n} is the outward unit normal vector to $\partial B(t)$ and \mathbf{v} is the velocity at the boundary points.

4. THE PHILIP-DE VRIES MODEL FOR THERMALLY INDUCED MOISTURE MOVEMENT

The extension to a nonisothermal system describing heat transfer and moisture transport in porous materials under combined moisture and temperature gradients has been made by Philip and de Vries [18]. The system of partial differential equations describing this model assumes the following form:

$$\frac{\partial \theta}{\partial t} = \nabla \cdot (D_T \nabla T) + \nabla \cdot (D_\theta \nabla \theta) + \frac{\partial K_\theta}{\partial z}, \quad (15)$$

$$\rho c \frac{\partial T}{\partial t} = \nabla \cdot (\lambda \nabla T) - \rho L \nabla \cdot (D_{\theta_{\text{vap}}} \nabla \theta), \quad (16)$$

where θ is the volumetric moisture content; T is the temperature; ∇ is the gradient operator. The parameters governing the model are: the thermal moisture diffusivity D_T ; the unsaturated hydraulic conductivity K_θ ; the isothermal moisture diffusivity D_θ ; the volumetric heat capacity of the porous medium ρc ; the thermal conductivity λ ; the latent heat of vaporization L and the isothermal vapour diffusivity $D_{\theta_{\text{vap}}}$. As was pointed out by Philip and de Vries, the hydro-thermal parameters D_T , D_θ , $D_{\theta_{\text{vap}}}$ and λ are generally θ - and T -dependent. On the other hand, these authors have not indicated the nature of the functional representations of these parameters. In subsequent studies Philip [19–21] concluded that thermal effects arising from the heat of wetting have negligible influence on the processes such as absorption or infiltration. In the opinion of the authors this assertion is true if the considered domain is either

semi-infinitely bounded or unbounded and where appropriate radiation conditions are operative. Indeed, in that case material parameters are predominantly moisture dependent since the heat dissipation occurs much faster than the moisture transport process: thermal and moisture transport processes become independent of each other.

The determination of the functional forms for D_θ , D_T , etc. applicable to porous geomaterials has been the subject of recent research (see e.g. Yong and Xu [25], and Selvadurai [4]). For the bentonitic clay buffer materials that have been proposed for use in the nuclear waste disposal scheme, Selvadurai [4] has developed the following functional relationships:

$$\left. \begin{aligned} D_\theta(T, \theta) &= \{0.0122 + 0.00132T\}e^{0.104\theta} \times 10^{-8} \text{ (m}^2/\text{s)} \\ D_T(T, \theta) &= \{-0.108 + 0.0200T + 1.805\theta\} \times 10^{-11} \text{ (m}^2/\text{s K)} \\ D_{\theta_{\text{vap}}}(T, \theta) &= \{-0.216 + 0.040T + 3.610\theta\} \times 10^{-14} \text{ (m}^2/\text{s)} \\ \lambda(T, \theta) &= \{2.685 + 0.00205T - 0.215\theta\} \text{ (J/ms K)}. \end{aligned} \right\} \quad (17)$$

These representations have been derived by making use of experimental data on moisture redistribution occurring in one-dimensional column tests. For convenience we write (17) in the form:

$$\left. \begin{aligned} D_\theta(T, \theta) &= (\alpha_1 + \alpha_2 T)e^{\alpha_3 \theta}, \\ D_T(T, \theta) &= \alpha_4 + \alpha_5 T + \alpha_6 \theta \\ D_{\theta_{\text{vap}}}(T, \theta) &= \alpha_{10} D_T(T, \theta) \\ \lambda(T, \theta) &= \alpha_7 + \alpha_8 \theta + \alpha_9 T \end{aligned} \right\} \quad (18)$$

where $\alpha_i, i = 1, \dots, 10$, follow from (17).

It will be shown in the next section that the representation (18) together with the equation (15) lead to a nonlinear convection–diffusion equation which yields the new class of solutions describing the redistribution of the moisture within a buffer region as a result of stationary thermal gradients.

5. NONLINEAR CONVECTION–DIFFUSION EQUATION FOR THE BUFFER SYSTEM SUBJECTED TO A CONSTANT TEMPERATURE GRADIENT

Considering the experimental studies reported in the literature [3,4] it is reasonable to assume that in the steady state, the temperature gradient is constant across the annular region of porous buffer. This is a plausible assumption as it relates to the long term thermal response and is also instrumental in simplifying the ensuing analysis. In the section which follows we will construct the nonlinear convection–diffusion equation for a buffer region which possesses a cylindrical geometry. The analysis, however, will be given in a generalized form and can be applied to alternative coordinate systems. We express the equation (15) in its invariant form by taking into account the functional representations (18). The gravitational effects in equation (15) may be neglected, since the problem is formulated in relation to the central plane of a waste emplacement borehole, i.e. for horizontal systems in which the influences of moisture and temperature gradients are more important than gravitational effects. Consequently the equation governing volumetric moisture content can be expressed as follows:

$$\frac{\partial \theta}{\partial t} = \text{div}[D_T(T, \theta)\text{grad } T] + \text{div}[D_\theta(T, \theta)\text{grad } \theta]. \quad (19)$$

The two operators of $\text{div}(\cdot)$ in (19) will be expressed in the cylindrical coordinates system (r, ϕ, z) utilizing simultaneously the assumptions stated above. We have the following:

$$\begin{aligned} \text{div}[D_T(T, \theta)\text{grad } T] &= D_T(T, \theta)\text{div}(\text{grad } T) + \text{grad } T \cdot \text{grad}[D_T(T, \theta)] \\ &= D_T(T, \theta)\text{div } \mathbf{G}_T + \mathbf{G}_T \cdot \text{grad}[D_T(T, \theta)] \end{aligned}$$

where \mathbf{G}_T is a constant temperature gradient. From vector analysis [22] we know that $\text{div } \mathbf{G}_T = 0$. Using this connection and the functional forms for D_T defined by (18), we can show that

$$\text{div}[D_T(T, \theta)\text{grad } T]_{(r, \phi, z)} = -\alpha_6 |\mathbf{G}_T| \frac{\partial \theta}{\partial r} + \alpha_5 |\mathbf{G}_T|^2, \quad (20)$$

where α_5 and α_6 are constants given by relations (17)₂ and (18)₂ and $|\mathbf{G}_T|$ is the magnitude of the constant temperature gradient across the annular buffer.

An expression similar to (20) can also be obtained for the second operator, $\text{div}[D_\theta(T, \theta)\text{grad } \theta]$ of equation (19), however, for the time being it is convenient to keep it in a compact invariant form, in the cylindrical coordinate system. We have

$$\text{div}[D_\theta(T, \theta)\text{grad } \theta]_{(r, \phi, z)} = \frac{1}{r} \frac{\partial}{\partial r} \left[r D_\theta(T, \theta) \frac{\partial \theta}{\partial r} \right]. \quad (21)$$

Making use of relations (20) and (21) and substituting them into equation (19) we obtain the following equation governing the variation of the volumetric moisture content, in the porous buffer material:

$$\frac{\partial \theta}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left[r D_\theta(T, \theta) \frac{\partial \theta}{\partial r} \right] - \alpha_6 |\mathbf{G}_T| \frac{\partial \theta}{\partial r} + \alpha_5 |\mathbf{G}_T|^2 \quad (22)$$

where $D_\theta(T, \theta)$, the isothermal moisture diffusivity, is given by (17)₁ and (18)₁. Equation (22) is meaningful from the point of view of Physics and Engineering. First of all, it is a single differential equation, and not a system of equations, describing the process of redistribution of the volumetric moisture content in the buffer material with respect to time and space. Secondly, it is a nonlinear convection–diffusion equation, which can lead to certain new solutions. Furthermore, the occurrence of unstable solutions can result from those solutions which exhibits a degenerate structure. The equation displays dependency with respect to temperature, temperature gradient and moisture gradient. The form of this equation is convenient for the investigation of the long term behaviour since it is likely that the solutions admit the similarity approximation. Finally, this equation will be linked thermodynamically to the general mass balance law, equation (13), from which it will be concluded that moisture depletion from the buffer is possible and that a singular interface in the form of a boundary between the dry and the moist regions of the buffer material can exist.

6. THE MAIN THEOREM RELATED TO THE NONLINEAR CONVECTION–DIFFUSION EQUATION

Since the moisture migration takes place between a heated internal boundary and an impervious outer boundary of an annular buffer region the domain of the solutions for equation (22) is considered as $\mathcal{D} = \bigcup_{t \in J} \{t\} \times [R_0, R_1]$, where R_0 is the radius of the heated boundary, R_1 is the outer radius of the buffer region, and J is the time interval. Within this domain the initial and boundary conditions must be specified for the solution of equation (22). The initial condition is known *a priori*, i.e.

$$\theta(r, t = 0) = \theta_0 = \text{constant}, \quad R_0 \leq r \leq R_1. \quad (23)$$

Also, the boundary condition at $r = R_1$ is specified by virtue of the impermeability condition, i.e.

$$\left. \frac{\partial \theta(r, t)}{\partial r} \right|_{r=R_1} = 0, \quad t > 0. \quad (24)$$

For a unique solution of the differential equation, a second boundary condition should be

located at $r = R_0$. This boundary condition is not known *a priori*. As a result, the initial-boundary value problem is incomplete and as such it cannot be solved unless some experimentally derived information is used to supplement the boundary condition at the interior region or the requirements for completeness and existence of a solution is achieved through the additional condition. The main theorem of the paper is as follows.

THEOREM. For the cylindrical geometry of an internally heated annular buffer region, there exists a critical value of the temperature gradient $|\mathbf{G}_T^{\text{cr}}|$ for which the moisture gradient $\partial\theta/\partial r$ assumes a non-negative value in the neighbourhood of R_0 , i.e.

$$\left. \frac{\partial\theta}{\partial r} \right|_{r=R_0} \geq 0 \quad \text{for} \quad |\mathbf{G}_T| \geq |\mathbf{G}_T^{\text{cr}}|, \tag{25}$$

and satisfies the following relationship:

$$\begin{aligned} R_0 D_\theta(T, \theta) \left. \frac{\partial\theta}{\partial r} \right|_{r=R_0} &= -|\mathbf{G}_T| \left\{ \alpha_6 [R_1 \theta(R_1, t) - R_0 \theta(R_0, t)] - (R_1 - R_0) \left[\alpha_6 \theta(\xi_0) + \frac{1}{2} \alpha_5 |\mathbf{G}_T| (R_1 + R_0) \right] \right\} \end{aligned} \tag{26}$$

where ξ_0 is the number specified between R_0 and R_1 .

PROOF. We assume that ψ_θ is a measure of the volumetric moisture content in the buffer, and that there is no singular surface $S(t)$ occurring within the system. Then, according to the relationship (5), the global mass integral of the volumetric moisture content in the buffer may be expressed in the form:

$$\psi_\theta(B_{\text{buf}}(t)) = \int_{B_{\text{buf}}(t)} \theta(\mathbf{x}, t) \, dV. \tag{27}$$

Following the theorem related to the differentiation of a volume integral, equation (14), the relation (27) leads to the expression for the overall time dependent variation of the volumetric moisture content as

$$\begin{aligned} \frac{d\psi_\theta(B_{\text{buf}}(t))}{dt} &= \frac{d}{dt} \int_{B_{\text{buf}}(t)} \theta(\mathbf{x}, t) \, dV \\ &= \int_{B_{\text{buf}}(t)} \frac{\partial\theta(\mathbf{x}, t)}{\partial t} \, dV + \int_{\partial B_{\text{buf}}(t)} \theta(\mathbf{x}, t) \mathbf{v} \cdot \mathbf{n} \, dA. \end{aligned} \tag{28}$$

In the latter the velocity of the boundary points of the buffer is zero, hence

$$\frac{d\psi_\theta(B_{\text{buf}}(t))}{dt} = \int_{B_{\text{buf}}(t)} \frac{\partial\theta(\mathbf{x}, t)}{\partial t} \, dV, \tag{29}$$

which in the cylindrical coordinates becomes

$$\frac{d\psi_\theta(B_{\text{buf}}(t))}{dt} = 2\pi \int_{R_0}^{R_1} r \frac{\partial\theta(r, t)}{\partial t} \, dr. \tag{30}$$

Making use of equations (22) and (24) and integrating the right-hand side of (30) by parts we obtain the following:

$$\begin{aligned} \frac{d\psi_\theta(B_{\text{buf}}(t))}{dt} &= 2\pi \left[\frac{1}{2} \alpha_5 |\mathbf{G}_T|^2 (R_1^2 - R_0^2) - \alpha_6 |\mathbf{G}_T| \cdot [R_1 \theta(R_1, t) - R_0 \theta(R_0, t)] \right. \\ &\quad \left. + \alpha_6 |\mathbf{G}_T| \int_{R_0}^{R_1} \theta(r, t) \, dr - R_0 D_\theta(R_0) \left. \frac{\partial\theta}{\partial r} \right|_{r=R_0} \right]. \end{aligned} \tag{31}$$

On the other hand, from equation (13) we know that

$$\frac{d\psi_\theta(B_{\text{buf}}(t))}{dt} = 0. \tag{32}$$

Thus, from (31) and (32), the following relationship is obtained:

$$R_0 D_\theta(R_0) \frac{\partial \theta}{\partial r} \Big|_{r=R_0} = \frac{1}{2} \alpha_5 |\mathbf{G}_T|^2 (R_1^2 - R_0^2) - \alpha_6 |\mathbf{G}_T| [R_1 \theta(R_1, t) - R_0 \theta(R_0, t)] + \alpha_6 |\mathbf{G}_T| \int_{R_0}^{R_1} \theta(r, t) dr. \quad (33)$$

Since $\theta(r, t)$ is a smooth function we can apply the Mean Value Theorem to the last term of (33) which gives

$$\alpha_6 |\mathbf{G}_T| \int_{R_0}^{R_1} \theta(r, t) dr = \alpha_6 |\mathbf{G}_T| (R_1 - R_0) \theta(\xi_0) \quad (34)$$

where $R_0 < \xi_0 < R_1$. Introducing (34) into (33) we get

$$R_0 D_\theta(R_0) \frac{\partial \theta}{\partial r} \Big|_{r=R_0} = -|\mathbf{G}_T| \left\{ \alpha_6 [R_1 \theta(R_1, t) - R_0 \theta(R_0, t)] - (R_1 - R_0) \left[\alpha_6 \theta(\xi_0) + \frac{1}{2} \alpha_5 |\mathbf{G}_T| (R_1 + R_0) \right] \right\} \quad (35)$$

which is identical to the relation (26). At this point the second part of the theorem can be proved. Let us assume that the expression in the large bracket of (35) is non-positive, i.e.

$$\alpha_6 [R_1 \theta(R_1, t) - R_0 \theta(R_0, t)] - (R_1 - R_0) \left[\alpha_6 \theta(\xi_0) + \frac{1}{2} \alpha_5 |\mathbf{G}_T| (R_1 + R_0) \right] \leq 0. \quad (36)$$

Then, the question arises as to the magnitude of the temperature gradient which satisfies the inequality (36).

Simple algebraic manipulation leads to the following constraints for $|\mathbf{G}_T|$:

$$|\mathbf{G}_T| \geq \frac{2\alpha_6}{\alpha_5 (R_1^2 - R_0^2)} \{R_1 \theta(R_1, t) - R_0 \theta(R_0, t) - (R_1 - R_0) \theta(\xi_0)\}. \quad (37)$$

The equality sign gives $|\mathbf{G}_T^{cr}|$ the critical value of $|\mathbf{G}_T|$. Thus, if $|\mathbf{G}_T| > |\mathbf{G}_T^{cr}|$ the value of the derivative $\frac{\partial \theta}{\partial r} \Big|_{r=R_0}$ is non-negative, and this means that the relation (25) holds. At this point the proof of the Theorem is complete.

7. IMPLICATIONS OF THE MAIN THEOREM

(1) It is important to note that the global mass in the solutions plays a critical role in achieving the completeness of the initial-boundary-value problems related to the heat-induced moisture migration problem. The general mass balance law leads to the determination of an unknown boundary condition located at the internal surface of the annular porous buffer material. We have proved that this condition is time-dependent and consequently the problem should be treated as a free or moving surface boundary-value problem. It is also clear that the condition (26) is inseparable from the partial differential equation describing the evolution of volumetric moisture content in the buffer, equation (22). Alternatively, from a thermodynamic perspective the relation (26) and inequality (25) are complementary conditions to equation (22) in terms of well-posedness of the problem relevant to long term predictions.

(2) The inequality (25) is extremely important in the assessment of the overall migration of moisture in the buffer. It states that, depending on the temperature gradient and the geometry of the buffer, the $\theta(r, t)$ can be monotonically increasing in the region close to the

heater. This implies that in the vicinity of the heated internal boundary of the buffer material can dry out. The consequences of this moisture depletion on the generation of microcracks within the buffer region is a phenomenon that has been observed in experiments. If such cracks can impede the conduction of heat from the heat generating waste, such processes can lead to thermal instabilities of the system.

(3) If the inequality (25) is satisfied then the $\theta(r, t)$ will reach its maximum value at an internal point of the buffer, say $r_0(t)$, where $R_0 < r_0(t) \leq R_1$ and this “max” must satisfy the following inequalities:

$$\theta_0 \leq \theta_{\max}(r_0(t), t) \leq \theta_{\text{saturation}} \tag{38}$$

Since, with time $r_0(t)$ can move towards the outer impervious boundary of the buffer, the existence of the singular surface in the buffer is possible and this can cause moisture depletion from the buffer.

(4) It should be noted that the singularity in (37) for $R_0 \rightarrow R_1$ is removable since for such a physical situation $\theta(\xi_0(t))$, $\theta(R_0, t)$ and $\theta(R_1, t)$ are of the same order and the limit of the right-hand side of (37) is zero. Thus, for any \mathbf{G}_T we have $|\mathbf{G}_T| \geq 0$ which is always true (i.e. the magnitude of any temperature gradient is always non-negative). This limit implies that from (35) we obtain

$$\left[\frac{\partial \theta}{\partial r} \right]_{r=R_0} = \left[\frac{\partial \theta}{\partial r} \right]_{r=R_1} = 0; \quad \text{for } R_0 \rightarrow R_1. \tag{39}$$

These conditions are equivalent to the physical constraint that the total moisture is conserved within the porous medium at all times. Consequently, when the annular buffer becomes very thin, the total moisture can only be redistributed within the space.

(5) The proof of the existence of a single local maximum for $\theta(r, t)$ at $r_0(t)$ merits further investigation. This study could be attempted by the application of procedures identified by Matano [5]. If a single local maximum exists at $r_0(t)$ where $R_0 < r_0(t) < R_1$ for $t > 0$, the typical envelope of solutions of (22) will be similar to the pattern identified in Fig. 3.

(6) It should be noted that the equation (22) can change its character from a parabolic to a hyperbolic differential equation. This implies the possible occurrence of an unstable solution for certain choices of α_6 , α_5 , D_θ and \mathbf{G}_T .

8. CONCLUSIONS

In this paper it is shown that the equation governing the heat-induced moisture movement within the porous medium is described by a nonlinear convection diffusion type equation [equation (22)], which yields a new class of solutions. It should be noted that this equation displays dependence with respect to temperature, temperature gradient and moisture gradient. It has been also shown that from the point of view of the thermodynamics of the problem, the moisture movement within the sealed porous medium is governed by a driving force which is expressed in terms of the temperature gradient and supplemented by an additional relationship resulting from the second law of thermodynamics. This relationship provides the additional boundary condition which is unknown at the initial stage and is referred to an internal surface within the buffer.

The main theorem related to the nonlinear convection–diffusion equation, equation (22), has been formulated and rigorously proved by virtue of the general mass balance law of continuum thermodynamics. The consequences of this theorem are many-fold and have been analysed in Section 7. In particular it may be concluded that depending on the temperature gradient and the geometry of the buffer, $\theta(r, t)$ can be monotonically increasing in the region close to the heater which implies that in the vicinity of the heated internal boundary the buffer

material can dry out. This thermally induced drying can lead to the overall thermal instability of the system (i.e. thermal runaway conditions).

It has been also concluded that there are possible occurrences of unstable solutions for certain choices of material parameters α_5 , α_6 , D_θ and the temperature gradient \mathbf{G}_T , since the equation (22) can change its character from a parabolic to a hyperbolic differential equation. This property is very important from the practical point of view in terms of the adequacy of the buffer in the long term. This change in the character can be observed in other complex phenomena and in recent years considerable interest has been focused on this class of problems in Engineering and Physics [23, 24].

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