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**THEMPAL EFFECTS, CREEP AND NONLINEAR RESPONSE
OF CONCRETE REACTOR VESSELS**

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SUMMARY

. A new mathematical model for prediction of pore pressure and moisture transfer in concrete heated well beyond 100°C is outlined. The salient features of the model are: (1) the hypothesis that the pore space available to capillary water grows with increasing temperature as well as increasing pressure in excess of saturation pressure, and (2) the hypothesis that moisture permeability increases by two orders of magnitude when passing 100°C. Permeability below 100°C is controlled by migration of adsorbed water through gel-pore sized necks on passages through the material; these necks are lost above 100°C and viscosity then governs. The driving force of moisture transfer may be considered as the gradient of pore pressure, which is defined as pressure of vapor rather than liquid water if concrete is not saturated. Thermodynamic properties of water may be used to determine sorption isotherms in saturated concrete. The theory is the necessary first step in rationally predicting thermal stresses and deformations, and assessing the danger of explosive spalling. However, analysis of creep and nonlinear triaxial behavior is also needed for this purpose. A brief review of recent achievements in these subjects is also given.

INTRODUCTION

One important problem in predicting the effect of a postulated hypothetical core disruptive accident in a prestressed concrete reactor vessel serving as the energy absorption barrier is the migration of moisture in the rapidly heated concrete wall and the build-up of pore pressure within concrete. Of major concern is the question whether the thermal stress and the pore pressure can induce explosive spalling of concrete, a phenomenon which is known from fire testing. Relevant measurements are scant and none of them give direct information on the values of pore pressures. Most of the test data on heated concrete have been obtained in the course of fire research on concrete building structures [1,2,3]; these are characterized by relatively thin cross sections, which lose moisture rapidly, while for reactor vessels much more massive structures must be studied.

One purpose of this contribution, based on report [4], is to outline in a condensed form the major findings that have recently been obtained on this problem as part of an extensive long-range program on the constitutive modeling of concrete at Northwestern University. In full detail, these findings will be presented in a subsequent comprehensive paper prepared jointly with W. Thonguthai [4]. Some of the theoretical arguments leading to the present formulation have been suggested in Ref. 5.

The purpose of the conference presentation is to also review recent results on the constitutive relations for creep at elevated temperature, on the probabilistic aspects of the extrapolation of short-time test data on creep, and on nonlinear triaxial constitutive relations.

MOISTURE AND HEAT TRANSFER IN CONCRETE

In irreversible thermodynamics, the general approach to the coupled moisture and heat transfer in porous solids is to write the vector of the mass flux of moisture, \mathbf{J} , and the heat flux vector, \mathbf{q} , as a linear combination of the gradients of pore water content w and of temperature T [6,7,8]. This linear relation is given by a square (2×2) coefficient matrix. The off-diagonal coefficients represent cross effects such as the Soret flux of moisture and the Dufour flux of heat. Studies of test data have, however, indicated that these complete transfer relations are not requisite for modeling concrete and that simplified transfer relations are possible if w is replaced by pore pressure p as the driving

force of mass transfer; i.e.,

$$\mathbf{J} = -\frac{a}{g} \operatorname{grad} p \quad (1)$$

$$\mathbf{q} = -b \operatorname{grad} T \quad (2)$$

in which g = gravity acceleration, introduced strictly for convenience: a = permeability (of dimension m/sec), and b = heat conductivity.

Eq. (1) is of course the same as Darcy's law, which is normally limited to saturated porous materials. On the basis of studies of test data, it appears, however, that Eq. (1) may be extended to non-saturated concrete, provided that pore pressure p is interpreted as the pressure of vapor rather than the pressure of liquid (capillary) water in the pores of heated non-saturated concrete [9,10,11]. Compared to w or other possible choices for the variable to represent the driving force of moisture transfer, the choice of p is more convenient because it allows eliminating $\operatorname{grad} T$ from the equation for \mathbf{J} . A certain thermal moisture flux (Soret flux) is, nevertheless, included in Eq. (1) because $\operatorname{grad} p$ is a linear function of both $\operatorname{grad} w$ and $\operatorname{grad} T$, as is clear from the sorption relation $w = w(p, T)$.

The condition of conservation of mass must account for the fact that water which is chemically bound in hydrated cement is upon heating released into the pores of concrete. Thus,

$$\frac{\partial w}{\partial t} = -\operatorname{div} \mathbf{J} + \frac{\partial w_d}{\partial t} \quad (3)$$

in which w represents the free water content, i.e., the mass of all free (not chemically bound) water per m^3 of concrete, and w_d denotes the mass of free water that has been released into the pores by dehydration (liberation of chemically bound water). At temperatures below $100^\circ C$, w_d may be used to represent the reverse phenomenon, i.e., the loss of free water caused by hydration, in which case the increments of w_d are negative. Inclusion of w_d in Eq. (3) is essential.

The condition of heat balance may be written as

$$\rho C \frac{\partial T}{\partial t} - C_a \frac{\partial w}{\partial t} - C_w \mathbf{J} \cdot \operatorname{grad} T = -\operatorname{div} \mathbf{q} \quad (4)$$

in which ρ , C = mass density and isobaric heat capacity of concrete (per

kg of concrete) including its chemically combined water but excluding its free water; C_a = heat capacity of free water plus the heat of adsorption of adsorbed water layers on pore walls; C_w = isobaric heat capacity of bulk (liquid) water. Term $C_w \cdot \text{grad } T$ represents heat convection due to movement of water [7,8]; normally this term is negligible, but in rapid heating, as in a reactor accident, this might not be true. The heat capacity terms of Eq. (2) may be further expressed as

$$\rho C \frac{\partial T}{\partial t} = \rho_s C_s \frac{\partial T}{\partial t} - C_d \frac{\partial w_d}{\partial t} \quad (5)$$

$$C_a \frac{\partial w}{\partial t} = \frac{\partial}{\partial t} (w_c H) - C_{ad} \frac{\partial w_{ad}}{\partial t} \quad (6)$$

in which ρ_s , C_s = mass of solid microstructure excluding hydrate water (per m^3 of concrete); C_d = heat of dehydration, C_{ad} = heat of adsorption on pore walls; w_c = amount of capillary water (per m^3 of concrete) = $w - w_{ad}$; w_{ad} = amount of water adsorbed on pore walls. Further complication stems from heat capacity C_s which includes the heat of various chemical reactions that occur at various temperatures as concrete is heated. The heat of vaporization of water does not figure explicitly in these equations but it may be included under $\partial(w H)/\partial t$.

Despite the above complex picture, it seems that the distinction between various terms contributing to heat capacity is not too important for the development of pore pressure, making it possible to neglect C_a and consider C approximately as a fixed function of temperature.

The boundary conditions for the heat and moisture transfer at the surface are

$$\eta \cdot J = B_w (p_b - p_{en}) \quad (7)$$

$$\eta \cdot q = B_T (T_b - T_{en}) + C_w \eta \cdot J \quad (8)$$

where η is the unit outward normal of the surface; p_{en} and T_{en} are the partial pressure p and temperature in the adjacent atmosphere, and p_b , T_b = value of p and T just under the surface of concrete. Term $C_w \eta \cdot J$ represents the heat loss due to the latent heat of moisture vaporization at the surface. Conditions of a perfectly sealed or perfectly insulated surface are obtained as $B_w \rightarrow 0$ or $B_T \rightarrow 0$, and conditions of perfect moisture or heat transmission are obtained as $B_w \rightarrow \infty$ and $B_T \rightarrow \infty$.

HYGROTHERMAL PROPERTIES OF CONCRETE

From studies of experimental data it has been found that permeability, a , tremendously varies with temperature, which has profound effects on the development of pore pressure. The following equation, graphically represented in Fig. 1, has been found to give reasonable fits of test data:

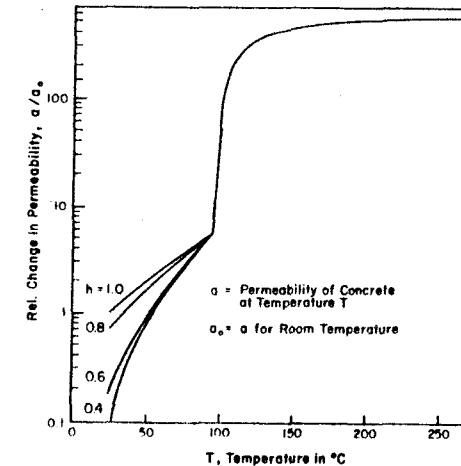


Fig. 1 Dependence of Moisture Permeability on Temperature and Relative Vapor Pressure

$$\text{For } T \leq 95^\circ\text{C}: a = a_0 f_1(h) f_2(T) \quad (9a)$$

$$\text{For } T > 95^\circ\text{C}: a = a'_0 f_3(T) \quad \text{with } a'_0 = a_0 f_2(95^\circ\text{C}) \quad (9b)$$

where a_0 = reference permeability at 25°C , and [9,10,11]

$$f_1(h) = \alpha_T + \frac{1 - \alpha_T}{1 + [4(1-h)]^4} \quad \text{for } h \leq 1 \quad (10a)$$

$$f_1(h) = 1 \quad \text{for } h > 1 \quad (10b)$$

in which $h = p/p_{sat}(T)$, $p_{sat}(T)$ = saturation vapor pressure, $\alpha_T = 0.05$ at 25°C and 1.0 at 95°C , while α_T varies roughly linearly between 25°C and 95°C ;

$$f_2(T) = \exp \left[\frac{Q}{R} \left(\frac{1}{T_{abs}} - \frac{1}{T_{abs_0}} \right) \right] \quad (T \leq 95^\circ\text{C}) \quad (11)$$

in which T_{abs} is the absolute temperature, Q = activation energy of low temperature moisture diffusion [9-11], R = gas constant;

$$f_3(T) = \exp\left(\frac{T - 95}{0.881 + 0.214(T - 95)}\right) \quad (T > 95^\circ\text{C}) \quad (12)$$

in which T = temperature in $^\circ\text{C}$.

Function $f_2(T)$ implies the moisture transfer below 95°C to be governed by activation energy and to consist of migration of water molecules along adsorbed water layers in cement gel. This agrees with function $f_1(h)$, which also reflects this mechanism, indicating a decrease in the rate of migration with a decreasing thickness of adsorbed layers.

Function $f_3(T)$ represents an upward jump of permeability by two orders of magnitude (about 200-times) when passing from 95°C to 105°C (Fig. 1) which represents one major finding of this work. This jump explains the well known fact that heated concrete can be dried orders of magnitude faster than concrete at normal temperatures.

To give a physical explanation of this permeability jump that is consistent with the adsorbed water migration at normal temperature, a new hypothesis is advanced. The rate of moisture transfer at ordinary temperature must be controlled by narrow "necks" on the transfer passages of smallest resistance through the material. These necks are of gel-pore dimensions, below about 10 molecules in thickness, so that water can pass through them only in adsorbed state, but not in vapor or liquid state which prevails along the rest of the passage. Upon heating beyond 95°C these necks are greatly widened, due to smoothening of the rough solid surfaces. Assuming the necks to be short and sparsely spaced along the passage, the large increase in neck width can be reconciled with the fact that the increase in the total pore volume upon heating is relatively modest. After the narrow necks are lost ($T > 105^\circ\text{C}$), the moisture transfer must be governed chiefly by the viscosity of the steam, which varies only mildly as temperature increases further. This is reflected in function $f_3(T)$. Moreover, when viscosity governs, permeability can no longer depend on pore pressure, as is reflected by the fact that $\alpha = 1$ for $T > 95^\circ\text{C}$.

Another property of major importance is the variation of that part of pore space which is available to capillary (liquid) water at pressures $p \geq p_{sat}(T)$. Obviously, the well-known thermodynamic properties of water

(as given, e.g., by ASME Steam Tables) must be applicable to capillary water. This would lead, however, to patently incorrect results if the pore were assumed to be constant and concrete were considered to be fully saturated before heating. One would then obtain pore pressures in excess of 1000 atm upon heating to 150°C , whereas the highest pore pressure ever recorded in heated concrete is about 8 atm. The fact that often concrete does not crack also implies the pore pressure to be relatively small.

Thus, it is necessary to assume that pore volume n available to capillary water significantly increases with both temperature and pore pressure. The following relations have been found to fit test data reasonably well:

$$w = (1 + 3\varepsilon^v)\frac{n}{v}, \quad n = \left(n_0 + \frac{w_d(T) - w_{d0}}{\rho_0}\right)p(h) \quad \text{for } h = \frac{p}{p_s} \geq 1.04 \quad (13)$$

$$p(h) = 1 + 0.12(h - 1.04). \quad (14)$$

Here n = capillary porosity (pore space available to capillary water within 1 m^3 of concrete), $v = v(T, p)$ = specific volume of water which depends on temperature and pressure as given by thermodynamic tables for water, ε^v = volumetric strain of concrete, $\rho_0 = 1 \text{ g/cm}^3$, $w_d(T) - w_{d0}$ = amount of free water released into the pores by dehydration = decrease of weight of chemically bound water from $T_0 = 25^\circ\text{C}$ to T , and n_0 = capillary porosity at 25°C . The sorption isotherms of saturated concrete based on Eqs. (13) and (14) are exhibited in Fig. 2.

The relationship of pore pressure, water content and temperature must further be specified for unsaturated concrete (below the critical point of water, 374°C). Under the assumption that pore geometry does not change, one may deduce, from Kelvin relation for capillary pressure and Laplace equation for capillary meniscus, that $w \sim h^{1/m(T)}$. This assumption is of course not true; however lacking a more sophisticated theory one can assume that a relationship of the same form holds even for pores of changing geometry, but with different coefficients, determined so as to give the best fit of test data. In this manner it has been found that

$$\frac{w}{c} = \left(\frac{w_1}{c} h\right)^{1/m(T)} \quad \text{for } h \leq 0.96 \quad (15)$$

$$m(T) = 1.04 - \frac{T'}{22.3 + T'}, \quad T' = \left(\frac{T + 10}{T_0 + 10}\right)^2 \quad (16)$$

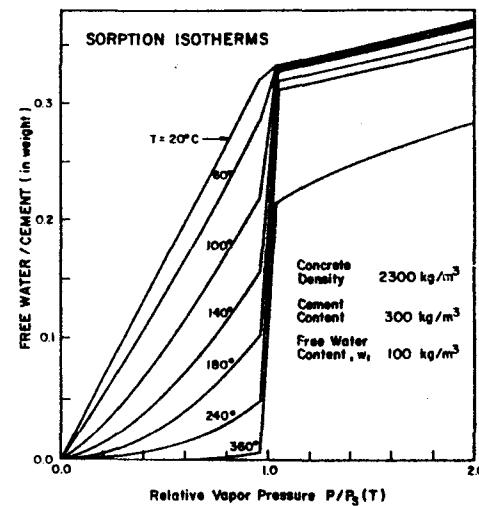


Fig. 2 Sorption Isotherms of Concrete at Various Temperatures

in which c = mass of anhydrous cement per m^3 of concrete, w_1 = saturation water content at 25°C.

Eqs. (13) and (14) are restricted to $h \geq 1.04$ and Eqs. (15) and (16) are restricted to $h \leq 0.96$. This is because a smooth transition is required between the saturated and nonsaturated regimes, as has been noticed when analyzing test data. The transition has been assumed as straight lines connecting the values of w at $h = 0.96$ and 1.04 at the same T . The complete isotherms as given by Eqs. (13)-(16) are shown in Fig. 2.

Further, less important, effects which ought to be included in prediction of pore pressure are the acceleration of aging by elevated temperatures below 100°C prior to the current time. The hydration acceleration is an effect that is opposite to dehydration w_d ; it also causes a significant rise in permeability. Roughly, $a_0 = a_1 10^x$, $x = \sqrt{a_2/t_e}$, $a_2 = \text{constant}$, $t_e = \text{equivalent hydration period (maturity)}$.

EXPERIMENTAL CALIBRATION

Although numerous test data have been accumulated in fire research, there seem to exist no direct measurements of sorption isotherms at

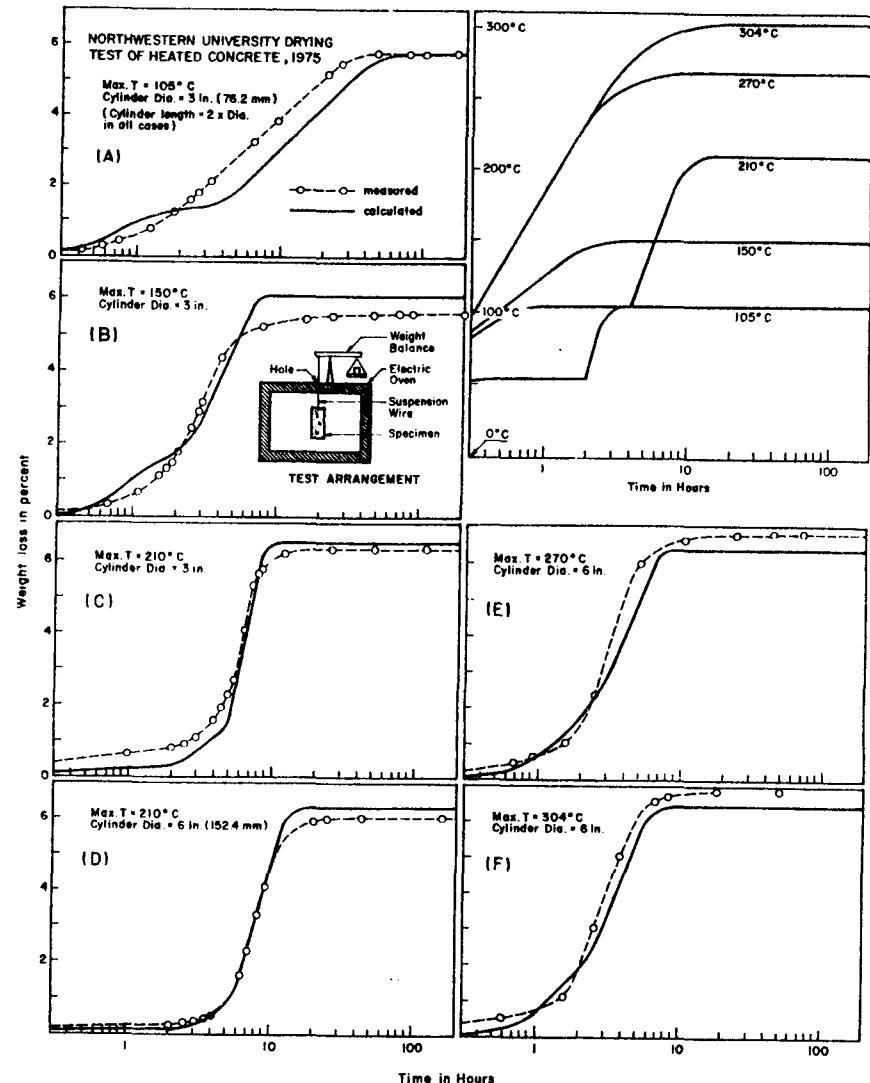


Fig. 3 Northwestern University Tests of Drying of Heated Concrete Cylinders and Theoretical Fits

elevated temperature and at pressures beyond the saturation pressure. In fact, even transient measurements that would provide indirect experimental information on pore pressures in heated concrete are extremely scant. Therefore, physical reasoning had to be substituted to a large extent to make up for the incompleteness of experimental evidence.

As no direct measurements seemed to exist on permeability of heated concrete either, drying tests have been conducted at Northwestern University (Fig. 3). Concrete cylinders of 3 inch (76 mm) and 6 inch (152 mm) diameters have been suspended in an oven and the rate of weight loss at various temperatures was measured. The measurements (Fig. 3) were then fitted by a finite element program based on the preceding formulation. These fits served mainly for calibration of the permeability dependence on temperature, and especially they revealed the sharp permeability jump around 100°C. (For details of test procedure as well as the finite element program, see [4].)

As far as calibration of the sorption isotherms is concerned, one needs mainly pore pressure measurements. From published test data, only those of England and Ross [12], and of Zhukov and Shevchenko [13,14] could be used. These data were fitted with the finite element program, and the fits achieved are shown in Figs. 4 and 5.

The finite element program used to fit the test data is based on Galerkin variational method applied to the field equations so as to reduce them to an ordinary matrix differential equation in time, which is then solved in time steps by Crank-Nicholson algorithm. A one-dimensional axisymmetric version has been used to fit the test data, but a two-dimensional axisymmetric version has also been developed. A complicating feature is that the linearized equation system obtained for each time-integration step is nonsymmetric.

MATHEMATICAL FORMULATION OF CREEP AND TRIAXIAL NONLINEAR DEFORMATION

For calculating stresses and deformations in heated concrete, creep is another major effect that must be taken into account. Existing rate-type models for creep at elevated temperature are suitable for computer analysis but the scope of their applicability beyond 100°C is not clear at present. Especially it is not known how concrete creeps when the escape of moisture within the time of heating is impossible, which is characteristic of the interior of a reactor vessel as well as of concrete

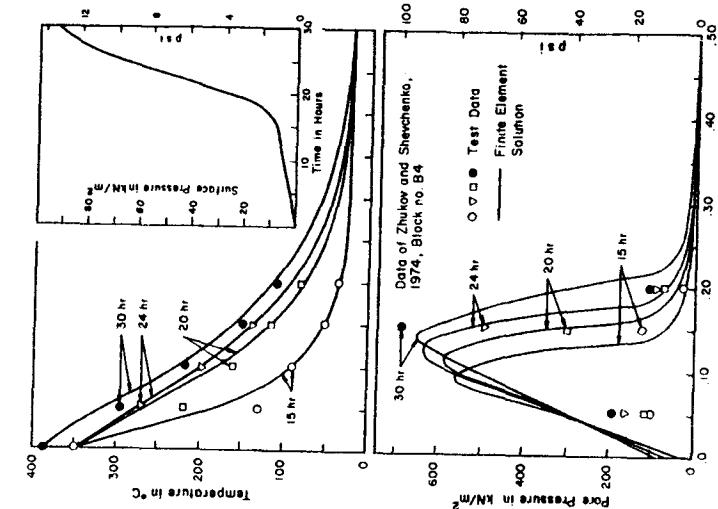


Fig. 5 Zhukov and Shevchenko's Measurements of Pore Pressure and Temperature in a Heated Concrete Block, and Theoretical Fits

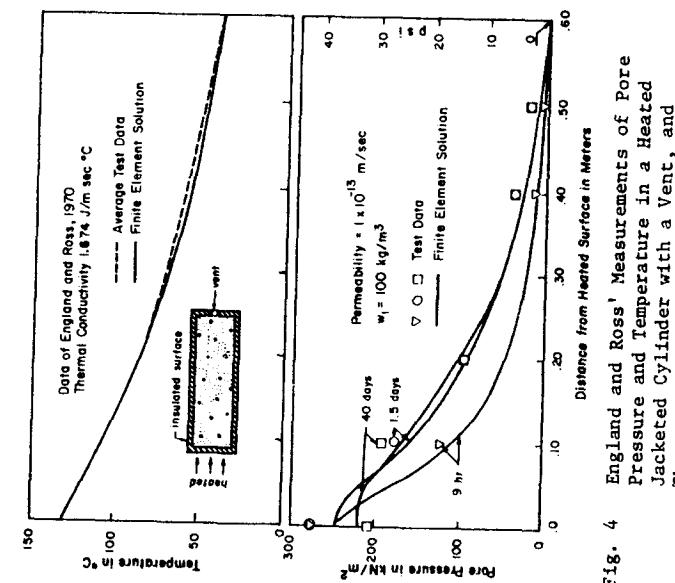


Fig. 4 England and Ross' Measurements of Pore Pressure and Temperature in a Heated Jacked Cylinder with a Vent, and Theoretical Fits

under a steel liner. By analogy with the behavior below 100°C, one might expect that in absence of a moisture loss the creep would be far less than during the moisture loss, although at the same time far higher than after the moisture loss. Creep data above 100°C are available, however, only for creep during and after moisture loss. A test program is under way at Northwestern University to determine creep of small cement paste cylinders which are jacketed and subjected in a triaxial device to a confining pressure which serves to eliminate jacket bulging and prevents thus any loss of moisture. Theoretical models under development represent a generalization of the existing rate-type model [11,15,16].

Another problem of practical concern for the design of reactor vessels or containments is how to extrapolate to long times (e.g., forty years) the short-time creep tests (say, of six months duration) carried out on the particular concrete to be used. Probabilistic aspect is an important element in such extrapolation, for the designer wants to know the 40-year creep value with a specified cut-off probability. For this purpose, a stochastic process model has been developed [17]. It represents creep as a non-stationary, local gamma process which can be transformed to a stationary gamma process. The model is based largely on statistical and physical arguments about the creep mechanism, but it has been also shown to agree reasonably well with various reported creep data. The mean value predictions of this process are compatible with the double power law for basic creep, which represents the best description of creep data available at present. The stochastic model [17] is limited to basic creep, but investigations are under way to extend it to conditions of variable water content and also to incorporate a physically more realistic model of aging [18].

Finally, prediction of response of reactor vessels and containments requires a constitutive model for nonlinear triaxial behavior at high stress. For this purpose, the endochronic formulation [19] appeared to be rather successful. However, an almost equally good model has more recently been obtained with an unconventional plasticity formulation [20,21,22] which includes fracturing terms of the type recently proposed by Dougill [23]. Comparison of endochronic and plastic approaches is given in [22], with particular attention to material stability and behavior at unloading, and a more general formulation combining the advantages of plastic and endochronic formulations is currently being developed.

CONCLUSION

Pore pressures and moisture migration in heated concrete can be predicted on a rational basis, utilizing thermodynamic properties of water. For this purpose it is necessary to account especially for the increase of pore space available to capillary water with increasing temperature and with pressure in excess of saturation pressure. A mathematical model for moisture transfer is necessary as the first step toward determining thermal stresses and deformations of heated concrete.

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