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TECHNICAL REPORT 83-13

AN INTEGRAL APPROACH TO RADIONUCLIDE TRANSPORT MODELLING IN FISSURED AND POROUS MEDIA

D. J. GILBY
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DECEMBER 1983

POLYDYNAMICS LTD, ZÜRICH

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A B S T R A C T

In order to fulfill the present and future requirements for the prediction of radionuclide migration in the geosphere a suite of numerical computer programs has been developed. They enable the transport of dissolved chemicals in general, of chains of radionuclides in particular and of heat to be calculated in either porous or fissured media.

The development of the conceptual models of the flow through porous continua and discrete fissures is related. This is followed by descriptions of the mechanisms affecting movement of the radionuclides and an overview of the resulting computer codes.

ZUSAMMENFASSUNG

Um die Anforderungen, die gegenwärtig und künftig an Vorhersagen über die Ausbreitung von Radionukliden in der Geosphäre gestellt werden, erfüllen zu können, wurde eine Reihe geeigneter Computerprogramme entwickelt. Sie ermöglichen die Berechnung des Transports chemischer Stoffe im allgemeinen, und insbesondere Zerfallsketten von Radionukliden sowie Wärme durch ein poröses oder zerklüftetes Medium.

Die Entwicklung der Modelle für die Strömung durch poröse Medien und einzelne Klüfte wird anschaulich dargestellt. Darauf folgen Beschreibungen der die Wanderung von Radionukliden beeinflussenden Mechanismen und ein Ueberblick über die entsprechenden Computerprogramme.

R E S U M E

Pour pouvoir satisfaire à la demande présente et future dans le domaine des prédictions de la migration des radionucléides dans la géosphère, une suite de programmes numériques pour calculatrice a été développée. Ces programmes permettent de calculer de façon générale le transport des produits chimiques dissous, de façon particulière le transport des chaines de radionucléides et enfin le transport de la chaleur, que ce soit en milieu poreux ou fissuré.

Le développement des modèles conceptuels d'écoulement au travers de milieux continus poreux et de fissures en nombres délimités est présenté. Suit une description des méchanismes affectant le mouvement des radionucléides ainsi qu'une revue des programmes de computer en résultant.

AN INTEGRAL APPROACH TO RADIONUCLIDE TRANSPORT MODELLING IN FISSURED AND POROUS MEDIA

1. INTRODUCTION

Disposal of radioactive waste by emplacement in deep and stable geologic formations is now under active consideration. A vital part of the assessment of feasibility of a waste repository is the prediction of the time required for radioactive material to reach the biosphere following any release from the repository to the geosphere. Clearly, the longer this transport process takes the lower will be the level of radioactivity delivered to the biosphere. Other important considerations include the location of the eventual release into the biosphere and the duration of release at significant activity levels.

The most probable mechanism by which radioactive material may be released from the repository and migrate to the biosphere is through groundwater contacting the waste, leaching out radionuclides and transporting them to water bodies used by man. Modelling the migration of radionuclides with groundwater is therefore essential in order to assess the potential radiological consequences of the geologic disposal option.

The purpose of this report is to describe the philosophy underlying the TROUGH (Transport of Radioactive Outflows in Underground Hydrology) package, a group of radionuclide migration models developed by the authors on behalf of NAGRA for use in the design and evaluation of a nuclear waste repository. The report does not attempt to cover in full detail the mathematical background of the models. For this the reader is referred to the technical descriptions contained in the user manuals ([7], [9], [10]).

In performing model studies of a waste repository the total system may be divided conveniently into four principal components:

a) The very near field - the region extending up to a few metres around an individual waste container.

b) The near field - a larger zone encompassing the whole repository extending to a few hundreds of metres from its centre.

c) The local domain or far field - a zone extending for a few kilometres from the repository.

d) The regional domain - containing the entire set of possible migration routes from the repository to the biosphere and including all possible sources of flowing groundwater.

These four components are closely interrelated, forming a "nested set" and the analysis of each one requires the results from at least one of the others as a boundary condition. This concept is discussed further within the report together with the relationship of the migration model to other models.

In the next chapter the fundamental problem of modelling the movement of groundwater is discussed. Migration mechanisms occurring within the groundwater are examined in Chapter 3. The interrelation of the different model scales and the interaction with other models forms the subject of Chapter 4 and the final Chapter briefly describes the methods of solution of the mathematical formulation of the problem.

2. GROUNDWATER FLOW MODELLING AND CLASSIFICATION OF WATER BEARING MEDIA

2.1 General

As a starting point in developing models of radionuclide migration with groundwater it is necessary first to consider the types of material through which the groundwater moves. Among the many types of igneous and sedimentary rocks we can, from the point of view of flow modelling, identify two extreme classes of material:

- Impermeable rock containing discrete fissures through which water can flow.

- Porous rock in which the flow paths are so finely distributed throughout the medium that the material can be considered as a homogeneous continuum.

Figure 2.1 illustrates these two categories of material, together with a "hybrid" type of rock, a porous matrix containing a large fissure.

These two material categories demand rather different flow modelling concepts. For the porous medium, the continuum assumption enables us to write down a single partial differential equation linking hydraulic head and flow velocity throughout the domain. The mathematical description of flow through a system of interconnecting fissures, on the other hand, involves the simultaneous solution of a set of such equations, one for each fissure in the network, with conditions of mass continuity applied at each intersection of two or more fissures providing the links between the equations.

2.2 Flow in Porous Media

The mathematical description and analysis of flow in a porous medium is based on Darcy's law, which states that the fluid flux through a porous medium is directly proportional to the local gradient of hydrostatic pressure and effectively also of piezometric head. The flux Q in direction x may be expressed as:

$$Q = -k \frac{dp}{dx} \sim - k\rho g \frac{dh}{dx} \tag{2.1a}$$

where:

g	is the gravitational acceleration
h	is the piezometric head
p	is the static pressure
ρ	is the fluid density

The constant of proportionality, k, is the intrinsic hydraulic conductivity of the medium.

While k is a property of the medium, in a completely general (non-isothermal) case the permeability must be considered to be also a function of the viscosity of the fluid. In addition, the average fluid velocity, u, in the material pores is related to the fluid flux by the local porosity, ε, so that the expression becomes:

$$u = \frac{Q}{\varepsilon} = \frac{k\rho g}{\varepsilon\mu} \frac{dh}{dx} = K \frac{dh}{dx} \tag{2.1b}$$

K is the hydraulic conductivity of the liquid/solid system

In a multi-dimensional problem the groundwater flow field is resolved into velocity components in each of the coordinate directions. We are then able to insert the Darcy description of velocity in each direction into the equation of mass conservation to obtain a single equation in piezometric head. For a non-isothermal model we must consider the effect of buoyancy forces in providing a vertical momentum source. This is done using the Boussinesq approximation, which means that density changes are considered only insofar as they produce buoyancy forces, mass continuity calculations being based on a constant reference density.

In non-isotropic media we must also take account of the tensorial nature of the permeability. Thus, for example, a head gradient in the x-direction will give rise to movement not only in the x-direction but also in the perpendicular directions. This can be envisaged as the flow slowly spreading as it moves down the pressure gradient.

When these factors are taken into proper account the condition of mass conservation at every point leads to the governing porous medium flow equation, written in 3-dimensional rectangular cartesian coordinates with z-vertical, as follows:

$$S\frac{\partial h}{\partial t} = \frac{\partial}{\partial x}\left(K_{xx}\,M\frac{\partial h}{\partial x} + K_{xy}\,M\frac{\partial h}{\partial y} + K_{xz}\,M\left(\frac{\partial h}{\partial z} - R\right)\right)$$

$$+ \frac{\partial}{\partial y}\left(K_{yy}\,M\frac{\partial h}{\partial y} + K_{yx}\,M\frac{\partial h}{\partial x} + K_{yz}\,M\left(\frac{\partial h}{\partial z} - R\right)\right)$$

$$+ \frac{\partial}{\partial z}\left(K_{zz}\,M\left(\frac{\partial h}{\partial z} - R\right) + K_{zx}\,M\frac{\partial h}{\partial x} + K_{zy}\,M\frac{\partial h}{\partial y}\right) \qquad (2.2)$$

where:

h	is the piezometric head (potential) $\left(\frac{p}{\rho_r g} + z\right)$
K_{ij}	are the hydraulic conductivity tensor components at the reference temperature T_r
M	is the viscosity ratio $\frac{\mu_r}{\mu}$
R	is the "density deficit" $(1 - \frac{\rho}{\rho_r})$
ρ	is the fluid density at the local temperature and pressure
ρ_r	is a reference fluid density
S	is the "storativity" of the permeable medium
z	is the vertical distance from an arbitrary reference level

The detailed derivation of this equation can be found in any reference work on flow in porous media (e.g. Bear [1]).

The solution of the above partial differential equation, for an appropriate set of boundary conditions, results in a hydrostatic pressure head field. This, in turn, can be inserted into the Darcy equation to give the local components of the flow velocity vector.

2.3 Flow in Fissured Media

At the low flow velocities normally encountered in natural groundwater movement, the flow in individual fissures can also be described by Darcy's law. For planar fissures a two dimensional form of equation 2.2 would be required, taking proper account of the orientation of the fissure when including a buoyancy term. However, such planes can be idealised as a collection of "flow tubes" which enables the use of a one-dimensional flow model and results in a greatly simplified expression of the mass continuity condition at the intersections of fissures. In addition, this flow-tube representation is conceptually close to the phenomenon of channelling that is believed to occur in natural fissures, where flow is not uniformly distributed over the exposed surface.

For a fissure flow model therefore we consider a set of interrelated solutions to equation 2.1 with K now being the individual fissure conductivity. By considering the ideal case of a smooth opening of constant hydraulic diameter we can express K in terms of an "apparent fissure aperture, δ".

The expression for the Hagen-Poiseuille flow (derived from [18]) through a smooth, parallel-sided slot of this aperture is:

$$Q = \bar{u}\delta = \frac{g\delta^3}{12\nu} \cdot \frac{\partial h}{\partial x} \qquad (2.3)$$

where:

g is the acceleration due to gravity

ν is the fluid kinematic viscosity

$\bar{u}$ is the average flow velocity across the opening

Consequently, in this ideal case:

$$K = \frac{g\delta^2}{12\nu}$$

It must, however, be borne in mind that aperture, shape and surface roughness may vary considerably along a single flow tube and the values of these properties must therefore be averaged along the flow tube.

The method of solution of the system of equations describing flow in a network of interconnecting fissures is shown briefly in chapter 5. Ultimately, the solution yields values for the velocity, and hence mass flow, of water along each fissure in the network for a given set of boundary conditions.

The fissured medium flow model clearly requires detailed knowledge of the pattern of fissures, their lengths, apertures, intersection frequency and hydraulic conductivity in order to set up the system of equations to be solved. At present it is possible to observe only the orientation and frequency of occurrence of fractures by means of exploratory borings. Equivalent hydraulic apertures and continuity of extent of fissures pose more of a problem and can be inferred only approximately from local measurements of permeability.

In the absence of a fully deterministic description of the fissure system we must consider the possibility of defining a network of fissures by statistical means - that is to say by generating arrays of likely fissure planes whose characteristics correspond to those observed and inferred. Individual analysis of one of the networks thus generated will yield a particular flow field, one of many possible results given the probabilistic nature of the fissure network.

Analysis of large numbers of networks of fissures will produce results from which the most-likely and the most unfavourable flow patterns can be determined with a degree of reliability depending on the scatter of the primary data and the number of probabilistic networks analysed. In respect of the safety assessment of a nuclear waste repository the most unfavourable flow field is the one that results in the most rapid transport of radionuclides to the biosphere and it is this case that is of principal interest. However, it is important in interpreting the results to bear in mind the statistical nature of the data on which the analysis is based, giving adequate weighting to the implications of the high probability cases while appropriately discounting, but not completely ignoring, the low probability of extreme cases.

2.4 Porous Medium Representation of Large Fissure Systems

When considering a larger scale of problem, the far field or regional scale, or when the material contains large numbers of fissures per unit volume, the fissure network model becomes increasingly cumbersome. At some stage it therefore becomes desirable to represent the fissured rock as an "equivalent porous medium". (The porous medium model does, of course, also have a direct application where a true porous rock mass exists within the modelling domain).

It is clear that a mass of rock containing a very large number of fissures can be expected to be representable as a porous continuum. The question that must be answered is at what scale and under what restrictions does the continuum approximation become valid in the sense that it adequately represents the behaviour of the fissured zone.

The hydraulic behaviour of systems of interconnecting fissures has been thoroughly investigated by Long et al. [13] and Long [14] using computer generated fissure networks. By controlling the parameters (mean, variance, etc.), under which the random variables describing the members of networks are generated it is possible to create systems with widely differing characteristics. Following extensive studies of large numbers of networks the following general conclusions have been reached:

- The hydraulic behaviour of a fracture system in a fixed volume becomes more like that of a homogeneous anisotropic material as fracture density increases.

- Fracture systems with distributed orientations behave more like homogeneous material than do systems with uniform orientations.

- Fracture systems with distributed apertures behave less like homogeneous material than do systems with uniform apertures.

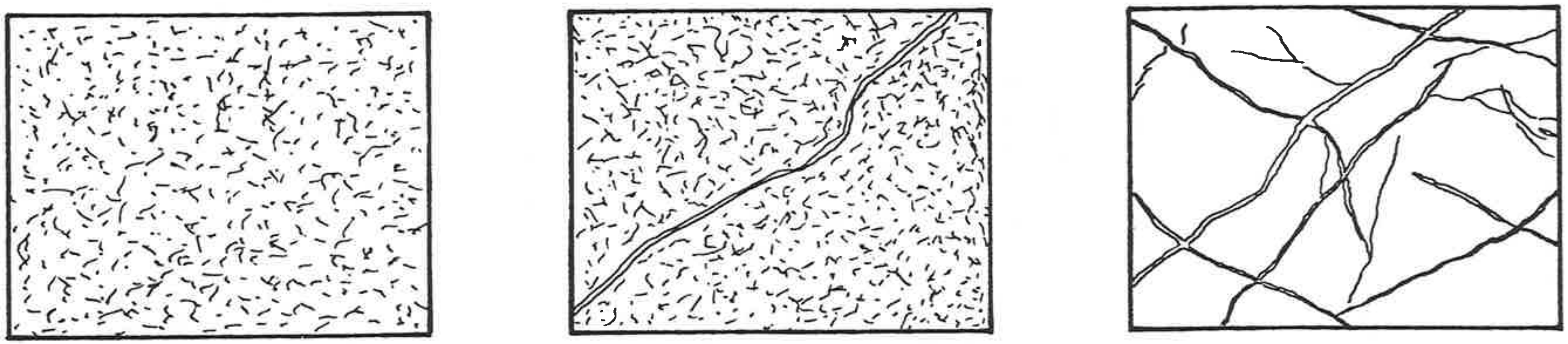
The first conclusion confirms the intuitive postulation that has already been made. The second and third conclusions require a little more discussion.

Naturally occurring fissures in rock can normally be grouped into families, all the members of one family having a similar orientation. Within a given sample of rock there will normally be only two or three families of fissures, although, sometimes there may be more, generated at different times and by different mechanisms. In directions aligned with the fissure planes of each family the conductivity will be relatively high while in other directions it may be orders of magnitude smaller. The irregularity of this directional dependence of hydraulic conductivity will become more pronounced as the amount of scatter within each family about the mean direction for that family is reduced, since the smaller directional scatter of fissures provides less opportunities for the flow to cross the domain in a direction not aligned with the mean fissure planes.

The tensorial nature of the porous medium permeability means that for an anisotropic medium the value of permeability changes smoothly with direction. Any significant deviation from this behaviour by a fissure network limits the validity of the equivalent porous medium treatment.

The explanation of the third conclusion derives from the fact that the conductivity of an individual fissure depends on the square of its apparent aperture, and the throughflow on the cube of the aperture. Consequently, when the aperture sizes are widely scattered there will be a small number with substantially larger apertures than the bulk of the family members. These carry a disproportionately large fraction of the flow and it will be the conductivity of these few extreme fissures that dominates the permeability of the entire fissure network. The overall appearance of the network permeability will therefore be closer to that of a system with considerably fewer fissures, that is, irregularly direction dependent.

All these factors must be considered when deciding between a discrete network model and a porous medium representation of the flow problem. Also, the two types of model have differing requirements for input data. The availability of reliable values for the many parameters involved in the equations must also influence the choice of the most appropriate approach.



Porous medium

Single fissure
in porous medium

Multiple fissures
(two families)

Figure 2.1: Sketches to illustrate the general material classes

3. MECHANISMS OF TRANSPORT IN MOVING GROUNDWATER

3.1 General

The flow models introduced in the previous chapter yield descriptions of the movement of groundwater through permeable media. We now turn to the problem of how radionuclides migrate with the moving groundwater.

The overall migration process is the net effect of several mechanisms, physical processes influenced principally by the movement of the water and the nature of the flow path, chemical processes involving reactions between the nuclides in the water and the solid material surrounding the flow area, and, in the case of radionuclides, the decay of the material being transported and the formation of new nuclides from a parent in a decay chain. This last process is of particular significance since the transport properties of the various members of a decay chain can differ substantially from each other, with the result that the transport of nuclides well down the chain is strongly influenced by that of more senior members. For example, an intrinsically slow moving nuclide could appear to move quite rapidly across the domain if it had a short lived, fast moving parent.

The several mechanisms involved in the migration process are discussed briefly in subsequent sections of this chapter.

3.2 Convection

Radionuclides carried along directly with the moving water are said to be convected. The convective flux of a nuclide can vary from place to place in a flow field as a result of changes in the velocity vector or the local concentration of nuclides. We can express mathematically the rate of accumulation of radionuclides at a point due to variation of the convective flux as follows:

$$- \frac{\partial}{\partial t} (\varepsilon C) = \frac{\partial}{\partial x} (\varepsilon u C) + \frac{\partial}{\partial y} (\varepsilon v C) + \frac{\partial}{\partial z} (\varepsilon w C) \qquad (3.1)$$

where:

C	is the concentration of radionuclides per unit volume,
u,v,w	are the components of the velocity vector in the direction of the coordinate axes x, y and z, while
ε	is the water-bearing void ratio or porosity of the host material.

The full 3-dimensional form of equation 3.1 is applicable to a porous medium. For fissure flow we would normally assume a uniform concentration distribution and zero velocity in a direction perpendicular to the plane of the fissure. Also, following the assumption of flow channels discussed in the previous chapter, the convective flux in a fissure flow model can be considered effectively 1-dimensional. In such a case the only ways in which the velocity can vary along a single flow tube are as a result of changes in cross-section or by leakage of water into (or out of) a surrounding porous matrix.

3.3 Diffusion and Dispersion

Diffusion is a molecular scale transport process that involves the migration of radionuclides (or any other scalar property such as thermal energy) away from local peaks of concentration. The process was first studied by Fick in 1855, who proposed a linear relationship between the diffusive flux and the concentration gradient, thus:

$$\dot{q}_A = -D \frac{dC_A}{dx} \tag{3.2}$$

where:

$\dot{q}_A$	is the flux of property A,
C_A	is the concentration of A and
D	is the diffusion coefficient.

This law of diffusion has found application in many different fields including heat transfer by conduction and momentum transfer by viscous forces. The effect is most clearly envisaged as a migration of concentration down a local concentration gradient.

An apparently analogous process, termed dispersion, takes place on a larger scale than the molecular when water moves through a porous or multiply fissured medium. In such a case the water follows a large number of separate flow paths across a domain, so that a parcel of nuclides injected instantaneously at one side will emerge over an extended period at the other. Furthermore, the concentration will also spread in a direction perpendicular to the general direction of flow since not all the possible flow paths will normally converge to a single point downstream from the injection point. Dispersion, then, has two components, longitudinal and transverse, both of which can be described mathematically by a law of the same form as Fick's, but with a much greater proportionality constant, the (longitudinal or transverse) dispersivity.

A considerable number of workers (see for instance: [2], [5], [12] and [20]) have investigated the phenomenon of diffusion and dispersion in porous media both theoretically and experimentally, their efforts being directed towards identifying some means of relating hydrodynamic dispersion to the flow conditions. The trend has been towards replacing the molecular diffusion coefficient of equation 3.2 with an effective coefficient of hydrodynamic dispersion which is a function of properties of the medium through which the water is flowing and the velocity of the flow. Including molecular diffusion in expressions for longitudinal and transverse dispersivity we can write, for unidirectional flow at a velocity u:

$$\begin{aligned} D_l &= a_l u + D_m \\ D_t &= a_t u + D_m \end{aligned} \qquad (3.3)$$

where:

D_m is the molecular diffusivity

D is the effective coefficient of dispersion

a is termed the dispersion length and is a property of the medium

l,t (subscripts) refer to longitudinal and transverse respectively

The situation becomes much more complicated in multi-dimensional flows since the dispersion coefficients are in fact functions not only of the magnitude of the velocity vector but also of its direction. Longitudinal dispersion takes place only along the direction of the velocity vector (which replaces u in 3.3) while transverse dispersion occurs in the plane perpendicular to that vector. The local velocity vector therefore defines a set of principal dispersion directions and fluxes. Resolving these fluxes into components in the three coordinate directions results in a tensorial form for the dispersivity. This means that the dispersive flux becomes a function not only of the concentration gradient in the direction of flux, but also of gradients in the orthogonal directions.

We therefore write the rate of accumulation of radionuclides due to dispersive fluxes as follows for two dimensions:

$$\frac{\partial}{\partial t}(C) = \frac{\partial}{\partial x}\left(D_{xx}\frac{\partial C}{\partial x} + D_{xy}\frac{\partial C}{\partial y}\right) + \frac{\partial}{\partial y}\left(D_{yx}\frac{\partial C}{\partial x} + D_{yy}\frac{\partial C}{\partial y}\right) \qquad (3.4)$$

It can be noted in passing that this expression is of the same form as equation 2.2 which could, therefore, be considered as the description of the diffusion of mechanical energy.

Equation 3.4 in its 2-dimensional form as shown and especially extended to 3-dimensions is valid only for a porous medium or a material so densely traversed by water-bearing fractures that it can be reasonably considered as an equivalent porous medium. The dramatic effects of dispersion in a porous continuum on the distribution of a transported nuclide has been shown by Schmocker [19]. Dispersion in a less well fissured medium would, like hydraulic permeability discussed in the previous chapter, tend to be irregularly direction dependent. In such a case the dispersion process would need to be modelled as an aggregation of the dispersion in each of the flow paths through the fissure system. A path could be two dimensional within a wide-open fissure, but using the flow tube analogy one needs to consider only the longitudinal effects. This leaves one-dimensional Fickian type diffusion, based on dispersivity modified to include the hydrodynamic dispersion generated in the flow tubes.

At the very slow flows normally encountered in groundwater migration through lightly fissured rock it could be reasonably argued that only molecular diffusion will have any significance in a single flow tube. A coarse scale mechanical dispersion will occur as the fluxes through individual tubes mix and separate while crossing the network. This process may be accounted for precisely in the fissured medium model as may be seen from the study by Schwartz et al. [21] using a statistically generated flowpath network. It is not relevant then in such a model to include hydrodynamic dispersion as conceived for a porous continuum. The mechanical dispersion engendered by the mixing/separation also outweighs molecular diffusion in most cases.

3.4 Radioactive Decay

For an individual member of a radioactive decay chain the balance between creation of new nuclides by decay of the parent, and the decay of that member of the chain can be expressed as follows:

$$\frac{\partial}{\partial t} C = S_\lambda = \lambda^* C^* - \lambda C \qquad (3.5)$$

where:

λ	is the radioactive decay constant
C	the number of nuclides present per unit volume
$*$	signifies the parent nuclide
S_λ	is the net rate of creation of nuclides

Depending on the number of nuclides of each species present and their respective decay constants the term S_λ can be either positive or negative at any instant, but must eventually become negative once the number of parent nuclides has decayed sufficiently.

The term S_λ appears as a specific part of a general source term in the overall radionuclide transport equation.

3.5 Chemical Sorption

Whenever a fluid containing a solute comes into contact with a new surface, ions can migrate out of the fluid and attach themselves to the solid surface. At the same time some ions will detach from the solid and return into solution in the liquid. The two processes of sorption and desorption will eventually come into a dynamic equilibrium, the two opposed fluxes balancing exactly. This state can be readily described by an equilibrium concentration ratio expressing the relationship between the concentration in the liquid and that in the solid (expressed on either a volumetric or an exposed surface area basis). For a linear isotherm, to which most data concerning the expected very low radionuclide concentrations are fitted, this ratio is independent of concentration. Any tendency for the concentration in the fluid to change will result in one or other of the two nuclide fluxes becoming greater, leading to a return to the equilibrium ratio at some new concentration level.

The consideration of the sorption process leads to the introduction of a new independent variable, the concentration of nuclides in the solid, into the transport equation. If we make the assumption that chemical equilibrium exists at all times between the solid and liquid the solid concentration can be replaced by a simple multiple of that in the liquid, thus eliminating the extra variable.

Since the nuclides sorbed on the solid do not migrate either by convection or diffusion the solid concentration appears in the transport equation only in the time dependent term and the decay source terms. We can therefore write the full transport equation for a single nuclide as follows:

$$\frac{\partial}{\partial t}(\varepsilon C_l) + \frac{\partial}{\partial t}\left[(1-\varepsilon)\rho_s C_s\right] + A_l = D_l - \lambda(\varepsilon C_l + (1-\varepsilon)\rho_s C_s)$$

$$+ \lambda^*(\varepsilon C_l^* + (1-\varepsilon)\rho_s C_s^*) \qquad (3.6)$$

where:

ε is the porosity of the medium,

A_l, D_l are the convective and diffusive flux gradient terms from equations 3.1 and 3.4, and

l, s (subscripts) refer to the liquid and solid concentrations respectively.

Conventionally, in determining the value of the equilibrium concentration ratio, C_l, the concentration in the liquid is expressed in mass units (or numbers of atoms) per unit volume of liquid while C_s is measured on a per unit mass basis. In consequence the definition of the equilibrium ratio, K_d is:

$$K_d = \frac{C_s}{C_l} \qquad (3.7)$$

We note that the group:

$$\varepsilon C_l + (1-\varepsilon)\rho_s C_s$$

appears repeatedly in equation 3.6 and introduce the definition 3.7 into this group to reduce it to:

$$\varepsilon\left(1 + \frac{1-\varepsilon}{\varepsilon}\rho_s K_d\right)C_l$$

The group of terms in brackets is clearly a property of the matrix material for a specific nuclide. It is usually termed the Retention or Retardation Factor, R, since it signifies the extent to which the mobile nuclides are held back by the sorption process.

Introducing this factor equation 3.6 becomes:

$$\frac{\partial}{\partial t}(\varepsilon R C_1) + A_1 = D_1 - \lambda \varepsilon R C_1 + \lambda^* \varepsilon R^* C_1^* \qquad (3.8)$$

There are, however, certain cases when the assumption of equilibrium will be invalid, for example in short duration field or laboratory experiments. In such cases, we must consider the solid concentration separately and formulate a term for the exchange flux between the solid and the liquid. At the same time we can write a separate differential equation governing the behaviour of the solid concentration, which is subject only to the processes of exchange with the liquid and radioactive decay. The exchange flux appears as a source/sink term in both of these equations, with opposite sign, and the second, solid concentration, equation must be solved simultaneously with the liquid concentration equation.

Unfortunately, at present little is known of the dynamics of the sorption process for radionuclides. Much investigation needs to be undertaken to identify a realistic model of the reaction and evaluate the parameters of that model.

3.6 Microfissure Diffusion

The concept of the fissure flow model is one of discrete flow paths separated by blocks of impermeable rock. Evidence is accumulating ([3], [4], [8], [15], [17]) to suggest that these blocks, although effectively impermeable to the main movement of groundwater, do contain water in micro-fissures and intergranular pores, which is accessible to mobile passing dissolved species. Any concentration of nuclides in the fluid flowing past such a block will be able to migrate into the solid material by molecular diffusion in the microfissure or pore water.

This process of micro-fissure diffusion can act as a significant delaying factor in the overall radionuclide migration. Nuclides diffusing into the solid remain there until the peak of concentration in the moving fissure has passed. They then diffuse back into the main migration path. Decay products formed from the originally trapped nuclides will also diffuse in the micro-fissures, generally with a different diffusivity from that of their parent. The net effect of the solid diffusion process is to reduce the magnitude of the concentration peak and extend the duration of the decline from that peak.

It has already been discussed how and why the fissure network which surrounds the solid blocks can only be described in some probabilistic way. A consequence of the statistical generation of the flow network is that the blocks of solid will be of irregular shape and size. We are thus presented with two alternatives in formulating a model of the matrix diffusion process:

- Model the diffusion in the individual irregularly shaped blocks using numerical methods.

- Idealise the blocks into a small number of simple shapes to permit the use of an analytic solution.

In the interests of computational economy we have chosen the latter course, idealising the rock blocks as slabs when considering long uninterrupted flow paths or spheres when several families of fissures intersect. Figure 3-1 depicts the idealisation of solid shapes in a small domain and shows that these ideal shapes can be of different sizes. In this way the idealisation can maintain the correct balance between solid-liquid interface area and diffusion path lengths. By allowing only these simple idealised shapes the solid diffusion process can be considered essentially 1-dimensional, permitting the use of relatively simple analytic solutions.

The idealised slab geometry is most applicable in a discrete fracture model while the spherical model is normally more appropriate when representing a large, well fissured region as an equivalent porous medium.

For the slightly simpler slab geometry, the equation solved for diffusion into the solid matrix is in terms of the distance ξ from the centre line of the slab towards the flowing water passing its face:

$$\frac{\partial^2 C_p}{\partial \xi^2} - \frac{1}{D}\frac{\partial C_p}{\partial t} = \frac{\lambda}{D} C_p - \frac{\lambda^*}{D}\frac{R_p^*}{R_p} C_p^* \qquad (3.9)$$

where:

D is the "apparent" diffusivity = D_p/R_p

p (suffix in C_p, D_p, R_p) signifies the pore water

If the slab is of thickness 21, the boundary and initial conditions for the solution are:

$C_p = \phi(t)$ at $\xi = \pm 1$

$C_p = 0$ at $t = 0$

Both opposing faces being supposed to be exposed to identical concentration and flow conditions. The symmetry of the situation permits an additional condition to be used, namely:

$\frac{\partial C_p}{\partial \xi} = 0$ at $\xi = 0$

Whatever the geometry, the analytic solution of the 1-dimensional diffusion equation, with appropriate extra terms to account for radioactive decay of the nuclides in the micro-fissures, can be manipulated to give an expression for the flux across the solid-fluid interface. This flux expression is incorporated into the transport equation as a source/sink term. Details of the derivation of the analytical solution used in the TROUGH models, and its incorporation into the numerical solution scheme have been published elsewhere ([6], [11]).

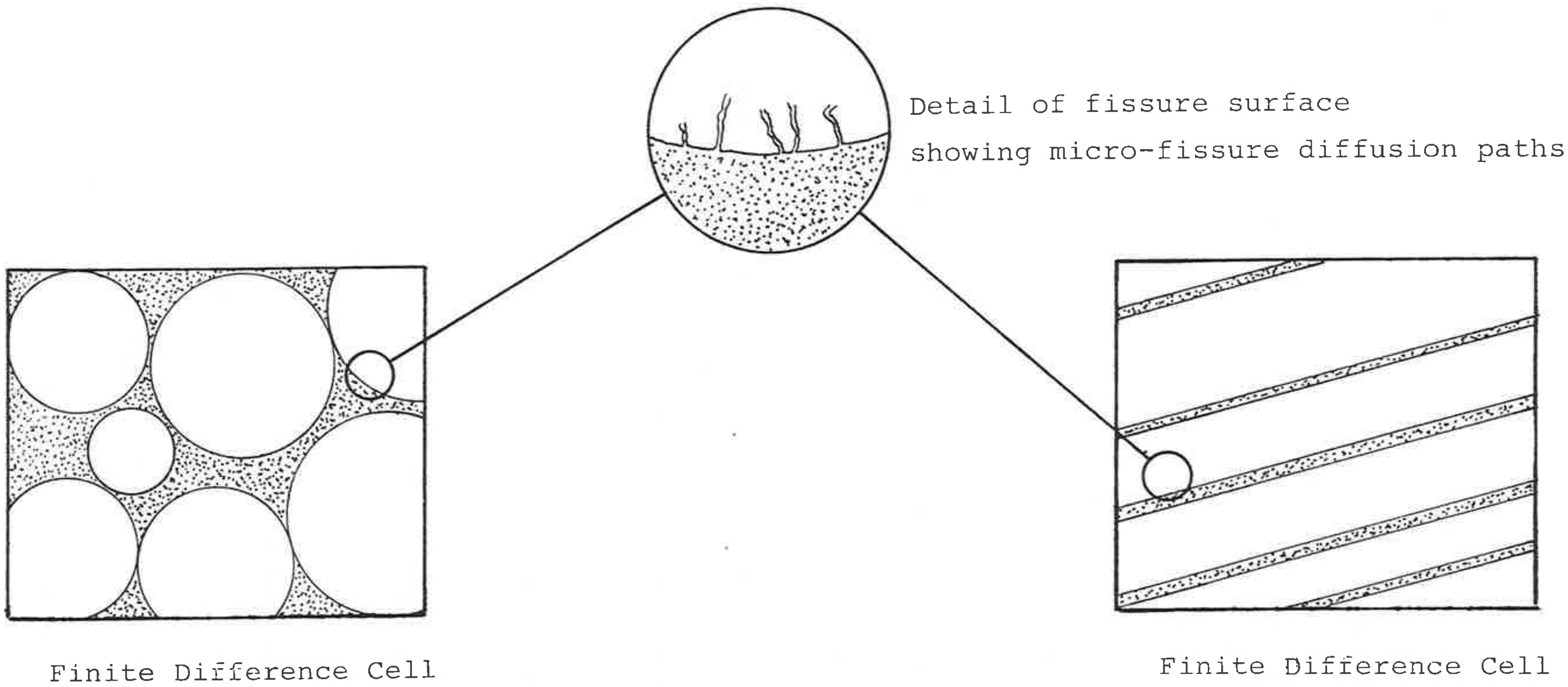


Fig. 3.1: Lumped parameter representation of solid blocks in a finite difference cell. Surface/volume ratio and diffusion path lengths may be adjusted by changing the number and characteristic dimensions of spheres or slabs.

4. INTERACTION WITH OTHER MODELS

Any mathematical model requires a minimum set of boundary conditions before it can be analysed to give a result. These boundary conditions may be dictated by the physical surroundings of the modelled region (for example, no groundwater flow across an impermeable rock layer along one or more edges) or they may be known values of the independent variable deriving from experimental investigations or from analysis on some other scale using a different model.

Within the context of the nuclear waste repository problem, two other major classes of model may interact with the TROUGH models by supplying boundary values of key variables.

On the large scale the groundwater flow model requires the specification of hydraulic heads or flow velocities along open boundaries of the model domain. Such data could come from investigations of groundwater recharge rates in the region being modelled, or from direct measurements of head in the water carrying zones at the edges of the domain. Alternatively, a larger scale model of regional groundwater movement could provide data on flow rates and heads around the boundaries of the zone that is to be modelled in detail.

Clearly, the flow model must be able to make use of these externally supplied boundary conditions in whichever form they appear.

At the other extreme of scale we need to know the rate at which radionuclides enter the groundwater from a waste container. This requires some detailed model of the process by which radionuclides are released from the waste material once the primary containment barrier, the canister containing the waste, has broken down.

The breakdown of this primary barrier and the subsequent leaching of nuclides from the waste material is an extremely complex process. In the first stage the corrosion of the container material by the groundwater and the influence on this of the chemical environment within the material immediately adjacent to the canister must be treated. At some point the nuclides will be able to diffuse through the corroded cladding. In this stage the process by which the nuclides escape from, or are leached from the waste material (probably in vitrified form), and the influence of the mixture of corroded canister material and surrounding matrix on both the leaching and diffusion of the nuclides must be modelled. Further, corrosion of the canister material, degradation of the waste, and changes in the chemistry of the surrounding material and the water it contains add to the complexity of the process in this stage. Much work is in progress at present to develop a fuller understanding of the true nature of the processes involved in breaking through the containment and leaching out the radionuclides. Ultimately, this work will lead to accurate near-field release models.

Possible alternative forms of the output of a near field model include a time history of the concentration of each nuclide in groundwater flowing past the point of release and a time history of the rate of release of radionuclides of each species at that point. A realistic model of the release process will also need to include some form of feedback from the transport model since the rate at which nuclides enter the groundwater could depend, to some extent, on the rate at which those released are carried away.

At very low flow rates the water in contact with the waste may become saturated with radionuclides; saturation occurs for many critical nuclides at extremely low concentrations. When it is reached, the extraction rate of the saturated species is radically slowed.

The insertion of the repository into the geosphere will, as mentioned above, result in a perturbation of the local geochemical situation. This means that the water chemistry in and immediately around the repository may change during a transient period which may be longer or shorter, depending upon the original situation (i.e. groundwater chemistry and mineralogy) and the materials used for the waste disposal. The effects may be felt as changes in solubility and sorption constants and eventually also in local variations in porosities and permeabilities.

To be fully effective therefore a release model must be either coupled with the transport model or include within itself a model of the migration and of the geochemistry in the very near field.

This leads us to the concept of flow and transport on models on different distance and time scales forming a nested set, each member of the set interacting with its neighbours in the next larger and smaller scales, in what must essentially be an iterative process.

Large scale models will be required to provide boundary condition data for the next smaller scale. Small scale models will provide specific information concerning events at some point or small area in the domain of a larger scale model.

Beginning with a regional flow model we "zoom in" in a series of steps to the very near field using a simple nuclide injection model in the intermediate steps. Following analysis of the very near field, including now a coupled leaching model, and with boundary conditions derived from the previous larger scale model, we can now reverse the procedure, if necessary back to the stage of modelling nuclide transport at the regional level. How far it is necessary to carry the process of re-expanding the scale of the model depends on the extent to which the initial nuclide injection model differs from the later more detailed model in the effect it has on predictions of concentration at larger distances from the point of injection.

5. IMPLEMENTATION

5.1 Overview

The previous chapters describe the way, in which the various radionuclide migration processes may be expressed mathematically. At this stage the migration model is, therefore, a set of differential equations in space and time with nuclide concentrations as the independent variables. In view of the many different processes involved, the time dependency of certain terms and the need to accommodate spatial variation of properties, an analytical solution of the overall set of equations is not a feasible possibility. To proceed we can therefore either develop analytical sub-models of the individual processes and attempt to combine their solutions in some way or we can search for numerical solutions of the equations.

The TROUGH family of codes are based on a method for the numerical solution of partial differential equations using a finite difference technique.

Three versions of TROUGH exist at present:

TROUGH 1-D
TROUGH 2-DP
TROUGH 2-DF

Version 1-D is used for studying chemical transport along predetermined paths, accounting for either equilibrium or kinetically dominated sorption processes, saturation and deposition and radioactive chain decay. For cases where the path passes through fractured rock, mass exchange with the blocks of rock and diffusion via the microfissure network within the solid matrix may be treated. These blocks may be treated as equivalent slabs or spheres.

Version 2-DP (two-dimensional, porous) permits the flow paths in a 2-dimensional field of any inclination to the vertical to be calculated, with or without the coupling of buoyancy effects due to temperature or large chemical concentrations. For this purpose, input data on heads, infiltration, outflows and well pumping rates may be used.

In addition to the flow calculations, the transport of heat through porous and quasi-porous continua and the same chemical processes as handled by the 1-D version can be treated. A particular feature is the treatment of the off-diagonal terms of the dispersion tensor.

Version 2-DF (two-dimensional, fissured) represents the alternative approach to transport through a fissured medium. Here a network of fissures is generated statistically and converted into a corresponding network of tube-like flowpaths. This concept enables what amount to one-dimensional solutions to be used for the transport phenomena.

Once again here the same transport phenomena may be treated for heat, stable chemical species or radionuclides.

5.2 Solution Technique

The modelled domain is divided into a number of control volumes (rectangular in two dimensions) for each of which a mass balance expression can be written in terms of concentrations in neighbouring control volumes. Figure 5.1 shows the computational grid of finite difference cells as used by TROUGH-1D. Figure 5.2 the same type of information for a porous continuum as may be modelled by TROUGH-2DP. Figure 5.3 shows a statistically generated network of flowpaths made with TROUGH-2DF.
The detailed view of each 1-dimensional network member is similar to Figure 5.1. It is assumed that the concentration varies linearly between the central nodes of adjacent control volumes so that the differential terms in the flux expressions reduce to simple algebraic differences. We are then left with a set of linear algebraic equations, the unknowns being the nodal concentrations. The solution of this set of equations yields a numerical description of the concentration distribution across the domain.

In a time dependent problem, fully implicit time differencing is used usually in both the 1-D and 2-DP codes. This means that the set of algebraic equations, including time dependent terms, are solved simultaneously in order to obtain a concentration field at the end of a finite time step. The implicit approach yields an unconditionally stable solution. A numerically much simpler approach is to determine the rate of change of concentration at each node at an instant in time and to extrapolate that rate of change forward for a finite period. This is termed explicit time differencing and although simpler, in that it does not necessitate simultaneous solution of the set of equations, it can result in numerical instabilities if the time steps are too large.

The requirements for a TROUGH-2DF solution are different. Here, for solution of the transport equations, time and distance steps are tied, so that a sharp front of chemical concentration may progress without any "numerical dispersion" and may hence remain sharp. In this case - negligible diffusion/dispersion in the transporting fluid - the explicit solution approach is preferred.

Throughout the whole family of codes advantage is taken of the similar formats of all transport equations. These are expressed in the conservation form and, where desirable, treated by a common solution method. The possibilities provided are summarized in Table 5-1.

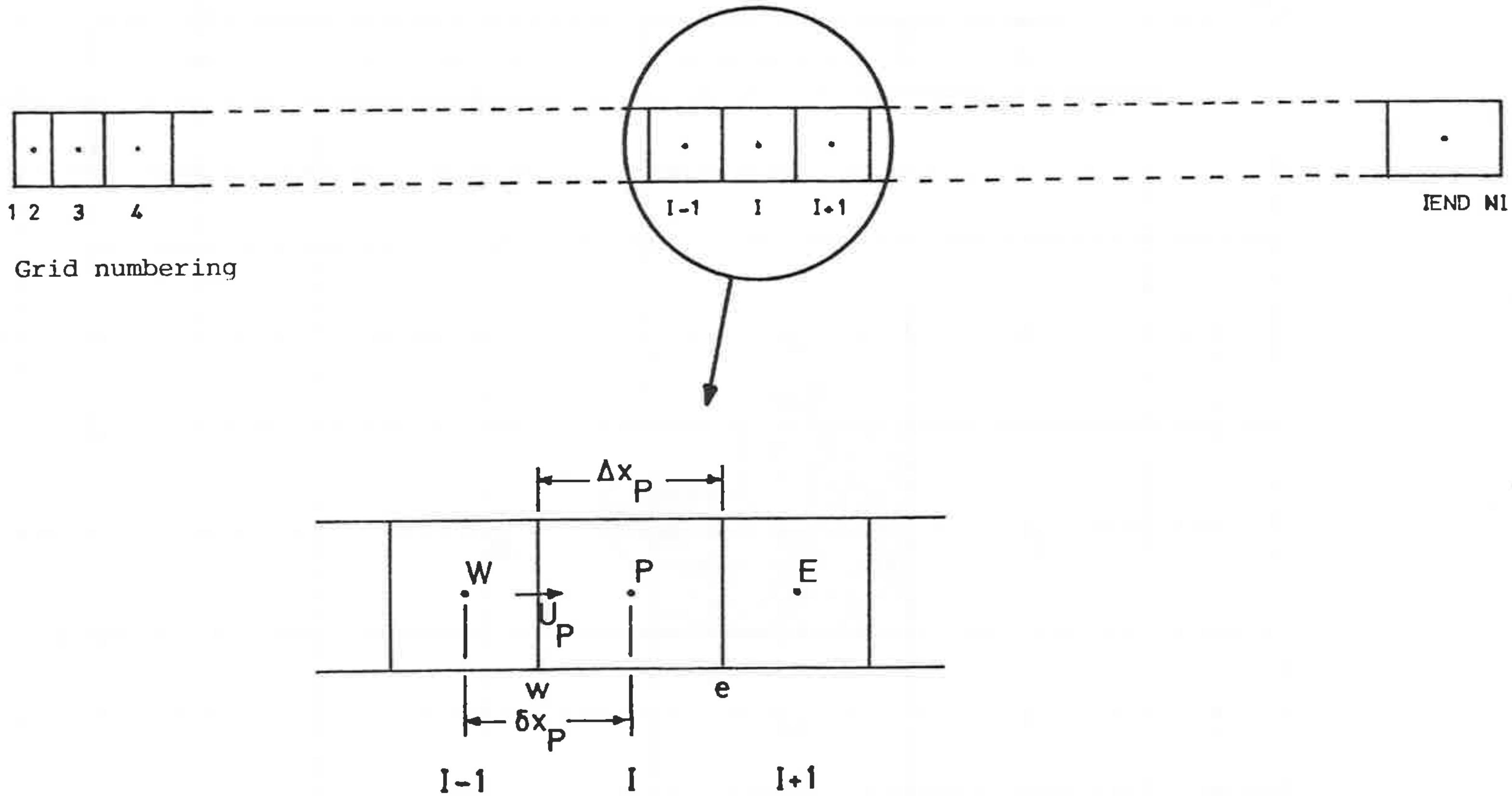
The codes are all described in greater detail in their respective user manuals.

5.3 Concluding Remarks

The computer programs briefly summarised in this chapter are tools for the detailed study of problems of flow and migration in both porous and fissured media. The host medium contains a mixture of pores and fissures and depending upon the physical scale in question, more or less detail is required in a model of the migration processes.

Verification of some aspects of the TROUGH codes has been possible in the framework of an international migration code comparison study, INTRACOIN, organised upon the initiative of the Swedish Nuclear Power Inspectorate. Reports are in preparation [22], [23] concerning this work.

Since the codes are being used both in Switzerland and abroad, new features are continually being added. The situation presented here then represents only the immediate situation.



Grid numbering

Enlargement of cell P and its neighbours

Figure 5-1: Layout and nomenclature for a 1-dimensional finite difference grid

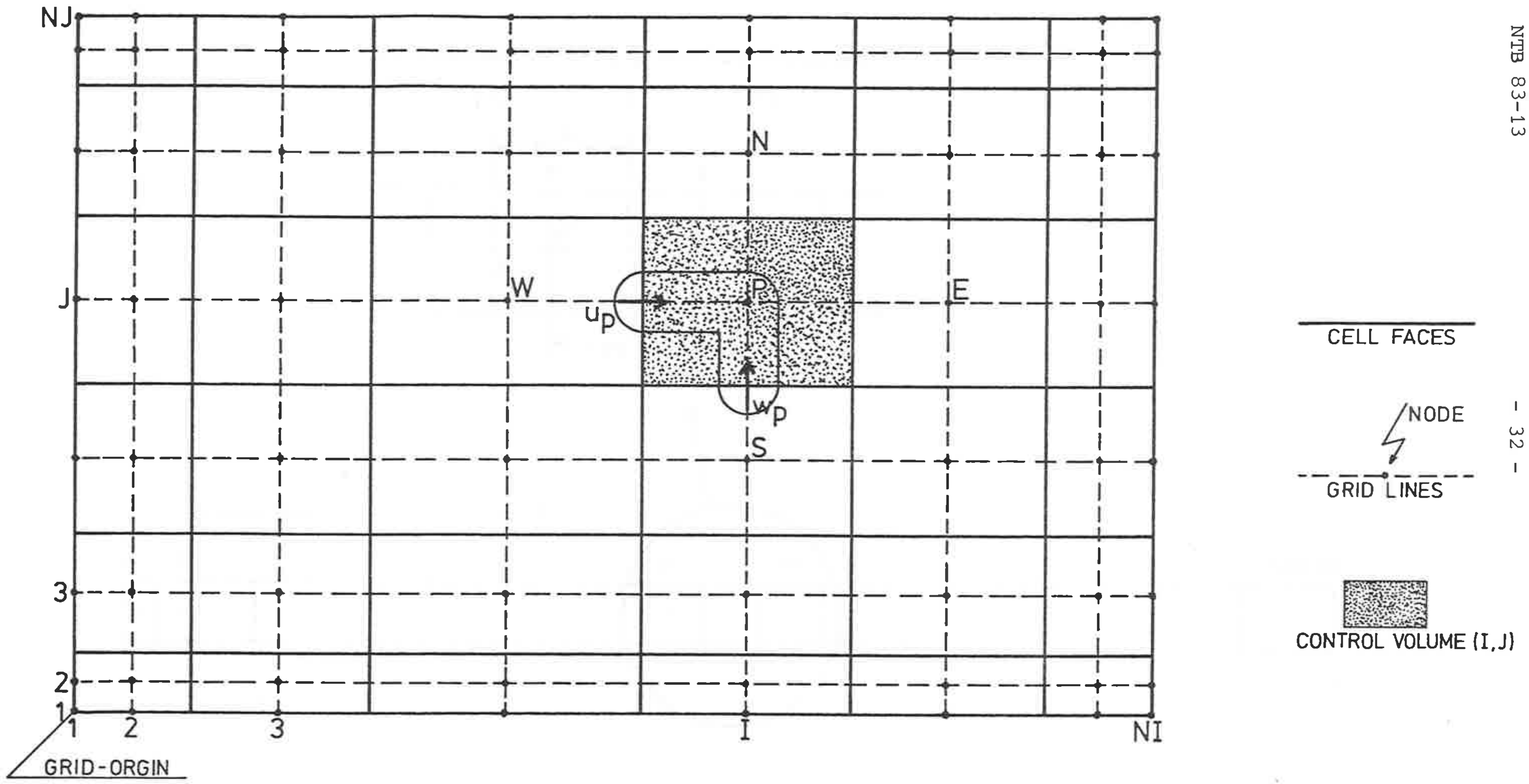


Figure 5-2: Layout and organisation of a rectangular grid for spatial discretisation of a 2-dimensional zone

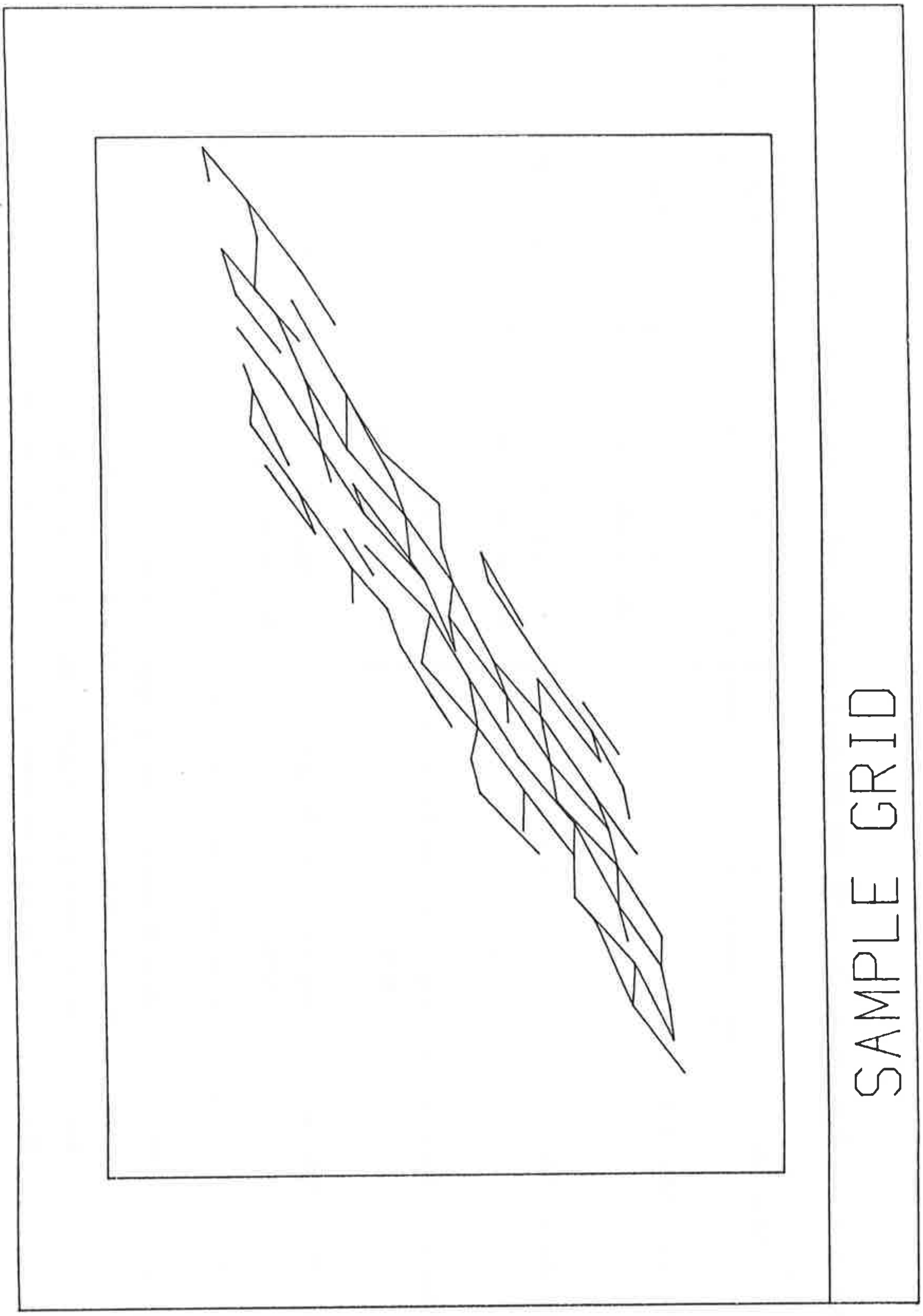


Figure 5-3: Statistically generated fissure network

Table 5-1: Overview of the different solution methods used in the TROUGH codes

Equation	1-D	2-DP	2-DF
Hydraulic head and flow	---	Implicit (BMA or ADI/TDMA)	BMA
Thermal energy	---	Implicit (BMA or ADI/TDMA)	Explicit
Chemical concentration	Implicit/C.N./Explicit (TDMA)	Implicit (BMA or ADI/TDMA)	Explicit
Nuclide concentration	Implicit/C.N./Explicit (TDMA)	Implicit (BMA or ADI/TDMA)	Explicit

Legend:

CN Crank Nicolson semi-implicit method

TDMA Tri-diagonal matrix algorithm with Gaussian elimination

BMA Banded matrix algorithm, using Gaussian elimination

ADI Alternating direction implicit, line-by-line/row-by-row

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