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# KBS TEKNISK RAPPORT

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DIFFUSION OF SOLUBLE MATERIALS  
IN A FLUID FILLING A POOUS  
MEDIUM

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AB Atomenergi 77-03-24

Objekt 11.05

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# Arbetsrapport

Diffusion of soluble materials in a fluid filling a  
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## Summary

The physical and mathematical foundations of diffusion in porous media are explained. Equations for diffusion of a radioactive nuclide chain are derived. The program DIWA for computing one-dimensional diffusion of one nuclide is described and results from such calculations are given. An underground deposition of wastes was assumed at a deep of 500 m. For a pessimistic assumption about the diffusion coefficient, no decay and no adsorption, the discharge at the ground level was negligible for at least 70000 years.

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## 1. Introduction

The problem of migration of radionuclides through an adsorbing medium has been investigated by D H Lester et al. [1]. In their work, the transport of the dissolved materials was caused by diffusion and by convection through a constant water flow. The emphasis was put upon the latter phenomenon and explicit equations were given for the case when there is no diffusion. In the present work the contrary assumption will be made, namely, that the only transport effect is due to diffusion. The physical background to this situation will be explained and the resulting expressions for the solution of the differential equations will be given in section 2. Section 3 contains a few comments about the physical constants involved and describes some numerical calculations for the simplest case, namely one-dimensional diffusion of only one type of nuclides.

2. Physical and mathematical considerations

The following notations will be used.

$c$	= fluid compressibility
$D$	= diffusion coefficient
$g$	= acceleration of gravity
$K$	= adsorption equilibrium constant
$K_p$	= permeability of a porous medium
$L$	= soil column length
$N_i$	= nuclide discharge rate
$N_i^0$	= nuclide initial inventory
$p$	= hydrostatic pressure
$Q$	= fluid source or sink
$t$	= time variable
$T$	= leach time
$V$	= fluid velocity for constant flow
$\bar{v}$	= mean flow rate per unit area (vector)
$x, y, z$	= Cartesian space coordinates
$\bar{e}_1, \bar{e}_2, \bar{e}_3$	= unit vectors in the directions of the space coordinates
$\varepsilon$	= porosity coefficient
$\lambda$	= decay constant
$\mu$	= viscosity
$\psi$	= flow potential
$\rho$	= density

The physical situation assumed is the following. Radionuclides dissolved in water are released from an underground deposition. The surrounding medium contains of a one-dimensional column of porous material and water adsorbed in this material. The ends of the column are at the ground surface and at the storage place. The solution of radionuclides is considered as a source. The transport of this fluid to the surface is caused by the following effects:

1. Thermal motion of the molecules.
2. Pressure differences between different points of the column.
3. Thermal convection.

It is easily seen that pressure differences can be derived from the effects 1 and 2 also. Therefore, it will hereafter be assumed that the motion is due to a pressure gradient.

The dynamic equilibrium between the forces on a fluid element in a porous medium is given by Darcy's law [2]. For a convenient description of this law, a flow potential,  $\psi$ , is introduced. It is given by the following expression:

$$\psi = g z + \int_{p_0}^p \frac{dp}{\rho} \quad (1)$$

where the first term on the r.h.s. is the gravitational potential and the second term is the hydrodynamic potential. The mean flow rate per unit area is then given by Darcy's law as

$$\bar{v} = - \frac{K_p \rho}{\mu} \nabla \psi \quad (2)$$

The transport equation is obtained by combining Eq (2) with the equation of continuity. Then,

$$\nabla \cdot \left( \frac{K_p \rho^2}{\mu} \nabla \psi \right) + Q = \varepsilon \frac{\partial \rho}{\partial t} \quad (3)$$

The system of Eqs (2) and (3) has been solved numerically by R W Nelson [6] for the pressure as an explicit variable.

In a mixture of two immiscible fluids, Eq (3) is valid for each of them separately.

It is now assumed that the compressibility of the fluid is constant, that is,

$$c = \frac{1}{\rho} \frac{d\rho}{dp} = \text{constant} \quad (4)$$

A combination of Eqs (1), (3) and (4) gives

$$\nabla \cdot \left[ \frac{K}{\mu} \left( \frac{1}{c} \nabla \rho + \bar{\alpha}_3 g \rho^2 \right) \right] + Q = \varepsilon \frac{\partial \rho}{\partial t} \quad (5)$$

If the pressure gradient is zero, the fluid has a constant velocity given by

$$V = - \frac{K \rho g}{\varepsilon \mu} \quad (6)$$

Therefore, if a constant velocity  $V$  can be superposed upon the flow rate caused by a pressure difference, the transport equation can be written

$$\nabla D \nabla \rho - V V \rho + \frac{Q}{\varepsilon} = \frac{\partial \rho}{\partial t} \quad (7)$$

where

$$D = \frac{K}{\varepsilon \mu c} \quad (8)$$

Equation (7) is the starting-point for the treatment in Ref 1 where the diffusion coefficient  $D$  was assumed to be constant and  $Q$  represents a source from radioactive decay and sinks due to adsorption and radioactive decay. In this work a still simpler model will be considered.

Firstly, the following special case of Eq (7) will be considered for a chain of radioactive nuclides:

$$\begin{aligned}
 D \frac{\partial^2 N_1}{\partial z^2} - K_1 \frac{\partial N_1}{\partial t} - K_1 \lambda_1 N_1 &= 0 \\
 D \frac{\partial^2 N_2}{\partial z^2} - K_2 \frac{\partial N_2}{\partial t} - K_2 \lambda_2 N_2 + K_1 \lambda_1 N_1 &= 0 \\
 \dots \dots \dots & \\
 D \frac{\partial^2 N_i}{\partial z^2} - K_i \frac{\partial N_i}{\partial t} - K_i \lambda_i N_i + K_{i-1} \lambda_{i-1} N_{i-1} &= 0
 \end{aligned}
 \tag{9}$$

This system of equations is identical with the transport equations in Ref 1 for  $V=0$ . The solutions for the general case are given by Eqs (2) to (22) in Ref 1. The functions  $F_n(i,j,\dots)$  are, however, changed. It is easy to obtain the corresponding functions for  $V=0$  if one observes that finite values will only be obtained for certain ratios of these functions. For example,  $F_{12}(i,j)$  will be infinite but this function occurs only as a member of the ratio

$$R = \frac{F_{12}(i,j)}{F_{17}(k,\ell)F_{17}(m,n)}
 \tag{10}$$

Thus, the corresponding functions were redefined so that the ratio  $R$  approaches the proper limit for  $V=0$ . This kind of redefinition is made for many functions but will not influence the final expressions for the solutions of Eq (9). The new functions will be written in terms of the following independent variables.

$$\eta = \frac{z}{L}
 \tag{11}$$

$$A_L = \frac{L^2}{Dt}
 \tag{12}$$

$$A_z = \frac{z^2}{Dt}
 \tag{13}$$

$$\tau_i = \lambda_i t
 \tag{14}$$

Then the functions needed for the solutions are

$$F_1(i) = N_1^0 \frac{\eta}{2t} \sqrt{\frac{K_i A_L}{\pi}} \exp\left(-\tau - \frac{K_i A_L}{4}\right) \quad (15)$$

$$F_2(i, j) = \frac{N_i^0}{2} \left( \frac{K_i \lambda_i}{K_i - K_j} \right) \quad (16)$$

$$F_3(i, j, k, \ell) = 0 \quad (17)$$

$$F_4(i, j, k, \ell) = \frac{K_i \tau_i - K_j \tau_j}{K_i - K_j} - \frac{K_i \tau_i - K_j \tau_j}{K_i - K_j} \quad (18)$$

$$F_5(i, j) = \exp\left(-\frac{K_i \tau_i - K_j \tau_j}{K_i - K_j}\right) \quad (19)$$

$$F_7(i, j) = \exp\left[\pm \eta \sqrt{\frac{4K_i K_j A_L (\tau_j - \tau_i)}{K_i - K_j}}\right] \quad (20)$$

$$F_9(i, j, k, \ell) = \operatorname{erfc} \frac{\eta}{2} \left[ \sqrt{\frac{K_i A_L}{K_i - K_j}} \pm \sqrt{\frac{K_j (\tau_j - \tau_i)}{K_i - K_j}} \right] \quad (21)$$

$$F_{10}(i) = \frac{N_i^0}{T} \exp(-\tau_i) \operatorname{erfc} \left( \frac{\eta}{2} \sqrt{K_i A_L} \right) \quad (22)$$

$$F_{11}(i, j) = \frac{N_i^0}{2T} \frac{\tau_i}{\tau_j - \tau_i} \quad (23)$$

$$F_{12}(i, j) = \frac{N_i^0}{2T} \tau_i \tau_j \quad (24)$$

$$F_{13}(i, j, k) = \frac{N_i^0}{2T} \frac{K_j \tau_i \tau_j}{K_j - K_k} \quad (25)$$

$$F_{14}(i, j, k, \ell) = \frac{N_i^0}{2T} \frac{K_i K_j \tau_i \tau_j}{(K_i - K_j)(K_j - K_k)} \quad (26)$$

$$F_{17}(i, j) = \tau_j - \tau_i \quad (27)$$

$$F_{18}(i, j, k) = \frac{K_j \tau_j - K_k \tau_k}{K_j - K_k} - \tau_i \quad (28)$$

$$F_{19}(i) = \exp(\tau_i) \tag{29}$$

$$F_{20}(i, j) = \exp\left[\pm n \sqrt{K_j A_L} (\tau_j - \tau_i)\right] \tag{30}$$

$$F_{21}(i) = \operatorname{erfc}\left(\frac{n}{2} \sqrt{K_i A_L}\right) \tag{31}$$

$$F_{22}(i) = F_{21}(i) \tag{32}$$

$$F_{23}(i, j) = \operatorname{erfc}\left(\frac{n}{2} \sqrt{K_j A_L} \pm \sqrt{\tau_j - \tau_i}\right) \tag{33}$$

$$F_{24}(i, j) = N_i^0 \frac{n}{2t} \sqrt{\frac{K_j A_L}{\pi}} \frac{\tau_i}{\tau_j - \tau_i} \left[ \exp(-\tau_i - K_j A_z) - \exp(-\tau_j - K_j A_z) \right] \tag{34}$$

$$F_{25}(i, j, k) = N_i^0 \left(\frac{1}{t}\right) \frac{\tau_i \tau_j K_i}{(K_k - K_i)(\tau_j - \tau_i)} \tag{35}$$

$$F_{26}(i, j) = \frac{N_i^0}{T} \frac{\lambda_i}{\lambda_j - \lambda_i} \left[ \exp(-\tau_i) - \exp(-\tau_j) \right] \operatorname{erfc}\left(\frac{n}{2} \sqrt{K_j A_L}\right) \tag{36}$$

$$F_{27}(i, j, k) = N_i^0 \frac{n}{2t} \sqrt{\frac{K_j A_L}{\pi}} \tau_i \tau_j \left[ \frac{\exp(-\tau_i)}{(\tau_j - \tau_i)(\tau_k - \tau_i)} + \frac{\exp(-\tau_j)}{(\tau_i - \tau_j)(\tau_k - \tau_j)} + \frac{\exp(-\tau_k)}{(\tau_i - \tau_k)(\tau_j - \tau_k)} \exp(-K_j A_z) \right] \tag{37}$$

$$F_{28}(i, j, k) = \frac{N_i^0}{T} \tau_i \tau_j \left[ \frac{\exp(-\tau_i)}{(\tau_j - \tau_i)(\tau_k - \tau_i)} + \frac{\exp(-\tau_j)}{(\tau_i - \tau_j)(\tau_k - \tau_j)} + \frac{\exp(-\tau_k)}{(\tau_i - \tau_k)(\tau_j - \tau_k)} \right] \operatorname{erfc}\left(\frac{n}{2} \sqrt{K_j A_L}\right) \tag{38}$$

The method of solution of Eq (9) is shown in the Appendix for the special case that  $i=3$  and  $K_1=K_2 \neq K_3$ . The same method can be applied for arbitrary values of relations between the adsorption constants  $K$ .

When only one nuclide is present the diffusion solution is given by Eq (15) for the impulse release. For decaying band release the proper combination of two expressions of the type given by Eq (22) gives the following solution:

$$N_{\text{band}} = \frac{N^0}{T} e^{-\lambda t} \left[ \operatorname{erfc}\left(\frac{z}{2} \sqrt{\frac{K}{Dt}}\right) - \operatorname{erfc}\left(\frac{z}{2} \sqrt{\frac{K}{D(t-T)}}\right) \right] \quad (39)$$

If  $t \leq T$ , only the first term within the bracket is to be used. It should be noted that the corresponding expression in Ref 1 is erroneous. It has not been recognized that the solution for  $t-T$  has different boundary conditions compared to the solution for time  $t$ .

### 3. Numerical computations of diffusion of a single isotope

The theory for one-dimensional diffusion of a single isotope is simple. The difficulty arises in determination of the diffusion constant for the actual material. Theoretically the diffusion constant is obtained from Eq (8). It is, however, more convenient to start with the bulk diffusion constant of the solute in the fluid,  $D_B$ . Then, for fluid-phase pore diffusion, the effective diffusion coefficient is [3], [4], [5]

$$D = \frac{D_B}{2} \quad (40)$$

It should be observed that the diffusion coefficient in Refs [3], [4] and [5] has a slightly different definition, so that the r.h.s. of Eq (7) should be multiplied by  $\epsilon$ . The factor 1/2 in Eq (40) is due to the increased path lengths in a porous medium compared to a bulk fluid.

The table below gives some experimental values of  $D_B$ , according to Wheeler [5].

Table 1

<u>Substance</u>	<u>T[°C]</u>	<u><math>D_B</math>[m<sup>2</sup>/s]</u>
H <sub>2</sub> in H <sub>2</sub> O	20	4 x10 <sup>-9</sup>
NaCl in H <sub>2</sub> O	20	1.4 x10 <sup>-9</sup>
Mannitol in H <sub>2</sub> O	70	1.56x10 <sup>-9</sup>

It seems obvious that for heavy particles in H<sub>2</sub>O,  $D_B$  is not larger than 4x10<sup>-9</sup> m<sup>2</sup>/s, that is 1.3x10<sup>-7</sup> km<sup>2</sup>/yr. This value has been used in the computations performed. Lester et al. [1] have, for unknown reasons, used 4.2x10<sup>-7</sup> km<sup>2</sup>/yr.

The porosity coefficient  $\epsilon$  which is present in Eqs (3) and (5) to (8), is given together with the permeability  $K_p$  in Table 2, according to Collins [2].

Table 2

<u>Substance</u>	<u><math>\epsilon</math></u>	<u><math>K_p</math> [Darcys]</u>
Silica powder	0.37-0.49	1.3x10 <sup>-2</sup> -5.1x10 <sup>-2</sup>
Loose sand	0.37-0.50	20-180
Soils	0.43-0.54	29-140
Sandstone	0.08-0.38	5x10 <sup>-4</sup> -3.0
Limestone	0.04-0.10	2x10 <sup>-4</sup> -4.5x10 <sup>-2</sup>
Brick	0.12-0.34	4.8x10 <sup>-3</sup> -2.2x10 <sup>-1</sup>

A computer program, DIWA, has been written for calculating the diffusion rate of a single isotope in a one-dimensional porous medium containing a fluid. The complementary error function is computed by the formula [7]

$$\operatorname{erfc}(x) = e^{-x^2} \sum_{n=1}^5 a_n t^n \quad (41)$$

where

$$\begin{aligned} a_1 &= 0.254829592 \\ a_2 &= 0.284496736 \\ a_3 &= 1.421413741 \\ a_4 &= -1.453152027 \\ a_5 &= 1.061405429 \end{aligned}$$

and

$$t = \frac{1}{1+px} \quad (42)$$

$$p = 0.3275911$$

The accuracy of this formula is  $1.5 \times 10^{-7}$ , but is not always sufficient for calculating the error functions in Eq (39) because the arguments often are very large. Then the difference between the both error functions is obtained from

$$\begin{aligned} \operatorname{erfc}(x_1) - \operatorname{erfc}(x_2) &= \frac{2}{\sqrt{\pi}} \int_{x_1}^{x_2} e^{-t^2} dt \approx \\ &\approx \frac{2}{\sqrt{\pi}} (x_2 - x_1) e^{-\left(\frac{x_1 + x_2}{2}\right)^2} \end{aligned} \quad (43)$$

if  $\left| \frac{x_2}{x_1} - 1 \right| < 10^{-7}$

The needed input data and their formats are given below. The variable notations are those used in the FORTRAN statements.

<u>Card</u>	<u>Position</u>	<u>Variable</u>	<u>Meaning</u>
1	1-12	CL	Soil column length, km
	13-24	TL	Leach time, yrs
	25-36	DO	Diffusion coefficient for bulk carrier, km <sup>2</sup> /yrs
	37-48	CNO	Nuclide initial inventory, C <sub>i</sub>
	49-60	DECAY	Decay const., inverted yrs
2	1-12	AK	Adsorption equilibrium const.
	13-24	DT	Time step, yrs
	25-36	DZ	Length step, km
3	1-6	NT	Number of time steps
	7-12	NZ	Number of length steps
	13-18	NBAND	0/1-impulse release/decaying band release.

Calculations have been made for a soil column length of 0.5 km. The nuclide release was either of impulse type or with a leach time of  $10^5$  yrs. The bulk diffusion coefficient was  $1.3 \times 10^{-7}$  km<sup>2</sup>/yr. The decay constant was zero or  $10^{-5}$  yrs<sup>-1</sup>. The adsorption equilibrium constant was 1.0 or 2.0. The nuclide initial inventory was 1.0. The results are shown in Figs 1-3. Fig 1 gives the discharge rate at the column end for impulse release.

Comparing the results with those of Lester et al., it is obvious that the contribution to leakage from diffusion is smaller than the contribution from convection by several orders of magnitude. The largest leakage occurs of course when  $K=1.0$  and  $\lambda=0$ . In this case the relative discharge at  $z=0.5$  km is less than  $10^{-11}$  for times smaller than 65000 years. At  $t=20000$  years, the value is only  $2.7 \times 10^{-24}$ . It is also very sensitive to the adsorption equilibrium constant. For  $K=2.0$  the leakage is already about 100 times smaller and it should be noted that the K-values given in Ref 1 vary between 100 and 50000.

Fig 2 shows the dependence of the discharge rate on the z-level. As could be expected this dependence is strong if the time is not very long. The conclusion is that each 100 meter deep difference in the position of the disposal site has a strong influence upon the discharge rate. On the other hand, comparing Figs 1 and 3 leads to the conclusion that only very long leach times have any influence upon the discharge rate. Thus, if diffusion is the only transport mechanism leach times less than 10000 years can be considered equivalent with impulse release.

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Solution of the diffusion transport equations for impulse release and two equal adsorption equilibrium constants

For impulse release the boundary conditions coupled to Eq (9) are [1]:

<u>Initial</u>	<u>Inlet</u>	<u>Outlet</u>
t=0, all z	t>0, z=0	t>0, z=∞
N <sub>1</sub> =0	N <sub>1</sub> =N <sub>1</sub> <sup>0</sup> δ(t)	N <sub>1</sub> =finite
N <sub>2</sub> =0	N <sub>2</sub> =N <sub>2</sub> <sup>0</sup> δ(t)	N <sub>2</sub> =finite
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N <sub>i</sub> =0	N <sub>i</sub> =N <sub>i</sub> <sup>0</sup> δ(t)	N <sub>i</sub> =finite

It will be assumed that i=3 . Applying the Laplace transform on Eq (9) leads to the following system of equations

$$\begin{aligned}
 D \frac{d^2 \bar{N}_1}{dz^2} - sK_1 \bar{N}_1 - \lambda_1 K_1 \bar{N}_1 &= 0 \\
 D \frac{d^2 \bar{N}_2}{dx^2} - sK_2 \bar{N}_2 - \lambda_2 K_2 \bar{N}_2 + \lambda_1 K_1 \bar{N}_1 &= 0 \\
 D \frac{d^2 \bar{N}_3}{dx^2} - sK_3 \bar{N}_3 - \lambda_3 K_3 \bar{N}_3 + \lambda_2 K_2 \bar{N}_2 &= 0
 \end{aligned}
 \tag{A1}$$

where

$$\bar{N}_i(z,s) = \int_0^\infty e^{-st} N_i(z,t) dt
 \tag{A2}$$

The boundary conditions coupled to Eq (A1) are

$$\bar{N}_i(0,s) = N_i^0
 \tag{A3}$$

The solution for  $\bar{N}_1$  can be obtained in a straight-forward way as

$$\bar{N}_1(z,s) = N_1^0 e^{-z \sqrt{\frac{K_1(\lambda_1+s)}{D}}}
 \tag{A4}$$

The inverse Laplace transform of Eq (A4) is well known (see for example M G Smith [8]) and gives the result

$$N_1(z,t) = \frac{N_1^0}{2t} \sqrt{\frac{K_1}{\pi Dt}} e^{-\lambda t - \frac{K_1 z^2}{4Dt}} \quad (A5)$$

Eq (A5) is of course identical with Eq (15). For solving the second row of Eq (A1), it is assumed that  $K_1=K_2$ . This gives a simple solution for  $N_2(z,t)$  and the general method will still be used when solving for  $N_3(z,t)$ . Inserting Eq (A4) into the second row of Eq (A1) gives

$$D \frac{d^2 \bar{N}_2}{dz^2} - (s+\lambda_2) K_1 \bar{N}_2 + \lambda_1 K_1 N_1^0 e^{-z \sqrt{\frac{K_1(\lambda_1+s)}{D}}} = 0 \quad (A6)$$

The solution of Eq (A6) with the proper boundary condition is

$$\begin{aligned} \bar{N}_2(z,s) = & \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 e^{-z \sqrt{\frac{K_1(\lambda_1+s)}{D}}} + \\ & + \left( N_2^0 - \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} \right) e^{-z \sqrt{\frac{K_1(\lambda_2+s)}{D}}} \end{aligned} \quad (A7)$$

The inverse transform of Eq (A7) [8] gives the final solution for  $N_2(z,t)$  as

$$\begin{aligned} N_2(z,t) = & \frac{1}{2t} \sqrt{\frac{K_1}{\pi Dt}} z e^{-\frac{K_1 z^2}{4Dt}} \left[ \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} e^{-\lambda_1 t} + \right. \\ & \left. + \left( N_2^0 - \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} \right) e^{-\lambda_2 t} \right] \end{aligned} \quad (A8)$$

Now Eq (A7) is inserted into the third row of Eq (A1). The differential equation for  $\bar{N}_3(z,s)$  then becomes

$$D \frac{d^2 \bar{N}_3}{dz^2} - (s+\lambda_3)K_3 \bar{N}_3 + \lambda_2 K_1 \left[ \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 e^{-z \sqrt{\frac{K_1(\lambda_1+s)}{D}}} + \left( N_2^0 - \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} \right) e^{-z \sqrt{\frac{K_1(\lambda_2+s)}{D}}} \right] = 0 \quad (A9)$$

The solution of Eq (A9) with the proper boundary conditions is

$$\bar{N}_3(z,s) = \frac{A_1}{s+a_1} e^{-z \sqrt{\frac{K_1(\lambda_1+s)}{D}}} + \frac{A_2}{s+a_2} e^{-z \sqrt{\frac{K_1(\lambda_2+s)}{D}}} + \left( N_3^0 - \frac{A_1}{s+a_1} - \frac{A_2}{s+a_2} \right) e^{-z \sqrt{\frac{K_3(\lambda_3+s)}{D}}} \quad (A10)$$

where

$$A_1 = \frac{\lambda_2 K_1}{K_3 - K_1} \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} \quad (A11)$$

$$a_1 = \frac{\lambda_3 K_3 - \lambda_1 K_1}{K_3 - K_1} \quad (A12)$$

$$A_2 = \frac{\lambda_2 K_2}{K_3 - K_2} \left( N_2^0 - \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} \right) \quad (A13)$$

$$a_2 = \frac{\lambda_3 K_3 - \lambda_2 K_2}{K_3 - K_2} \quad (A14)$$

and  $K_2 = K_1$

The inverse transform of Eq (A10) contains integrals of the following type:

$$F(a,b,c) = \frac{1}{2\pi i} \int_{-i\infty+\alpha}^{i\infty+\alpha} \frac{e^{st - \frac{\sqrt{s+b}}{c}}}{s+a} ds \quad (\text{A15})$$

where  $\alpha$  is such that the integration path goes to the right of all singularities and branch points. The convolution theorem gives

$$F(a,b,c) = \frac{e^{-bt}}{2\pi i} \int_{-i\infty+\alpha}^{i\infty+\alpha} \frac{e^{st - \frac{\sqrt{s}}{c}}}{s+a-b} ds \quad (\text{A16})$$

The solution of this integral is dependent on whether  $a-b$  is positive or negative. It is hereafter assumed that  $a-b$  is negative. The integral to be solved can then be written

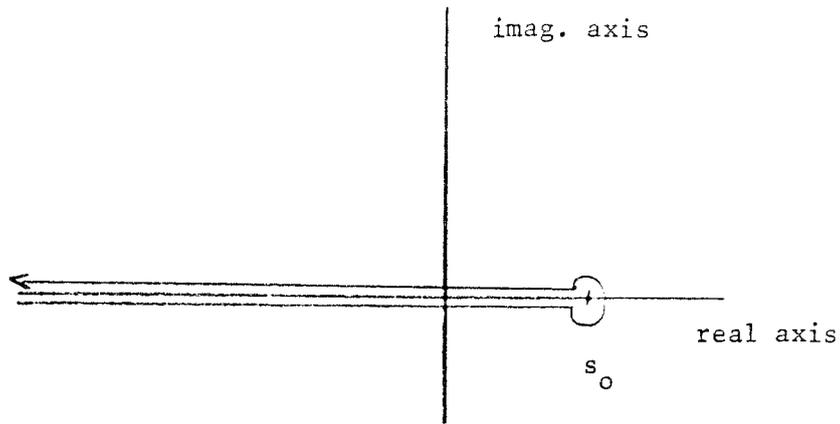
$$G(c,s_0) = \frac{1}{2\pi i} \int_L \frac{e^{st - \frac{\sqrt{s}}{c}}}{s-s_0} ds \quad (\text{A17})$$

where the positive constant  $s_0$  is

$$s_0 = \lambda_j + \frac{K_3(\lambda_i - \lambda_3)}{K_3 - K_i}, \quad \begin{matrix} i = 1, 2 \\ j = 1..3 \end{matrix} \quad (\text{A18})$$

$s_0$  is positive if  $\lambda_i - \lambda_3$  and  $K_3 - K_i$  have the same sign. Because only three nuclides are considered, it is meaningful to set  $\lambda_3 = 0$ . This is the same as assuming that all subsequent nuclides have the same K-values as the 3rd nuclide.

The integration path is determined by the branch line in the numerator and is shown in the figure below.



The branch line consists of the negative part of the real axis.  $s_0$  is a simple pole. The contributions between  $s=0$  and  $s=s_0$  cancel each other. Along the negative axis, the square root of the argument is

$$\sqrt{s} = \bar{i} \sqrt{|s|} \tag{A19}$$

Then, with  $\rho = s_0 - s$ ,

$$\begin{aligned} G(c, s_0) &= \frac{e^{s_0 t}}{2\pi i} \left\{ - \int_{s_0}^{\infty} \frac{e^{-\rho t} \left( e^{i \sqrt{\frac{\rho-s_0}{c}}} - e^{-i \sqrt{\frac{\rho-s_0}{c}}} \right)}{\rho} d\rho + \right. \\ &\quad \left. + i \int_{-\pi}^{\pi} e^{-\sqrt{\frac{s_0}{c}} \phi} d\phi \right\} = \tag{A20} \\ &= e^{s_0 t - \sqrt{\frac{s_0}{c}}} - \frac{1}{\pi} \int_0^{\infty} \frac{e^{-\rho t}}{\rho + s_0} \sin \sqrt{\frac{\rho}{c}} d\rho \end{aligned}$$

The integral in Eq (A20) can be solved by introducing a new variable  $\tau$  in the following way:

$$I = \int_0^{\infty} \frac{e^{-\rho t}}{\rho + s_0} \sin \sqrt{\frac{\rho}{c}} d\rho = e^{s_0 t} \int_t^{\infty} e^{-s_0 \tau} d\tau \int_0^{\infty} e^{-\tau \rho} \sin \sqrt{\frac{\rho}{c}} d\rho$$

(A21)

Setting  $\rho = r^2$  and integrating by parts gives

$$I = \frac{1}{\sqrt{c}} e^{s_0 t} \int_t^{\infty} e^{-s_0 \tau} \frac{d\tau}{\tau} \int_0^{\infty} e^{-\tau r^2} \cos\left(\frac{r}{c}\right) dr =$$

$$= \frac{1}{2} \sqrt{\frac{\pi}{c}} e^{s_0 t} \int_t^{\infty} e^{-s_0 \tau} \tau^{-\frac{3}{2}} \frac{1}{4c\tau} d\tau$$

(A22)

Then two transformations are made, namely

$$u = \tau^{-\frac{1}{2}}$$

(A23)

and

$$v = \frac{1}{2\sqrt{c}} u - \frac{\sqrt{s_0}}{u}$$

(A24)

whereby the integral becomes

$$I = \pi e^{s_0 t} \sqrt{\frac{s_0}{c}} \left[ 1 - \frac{1}{2} \operatorname{erfc}\left(\frac{1}{2\sqrt{ct}} - \sqrt{s_0 t}\right) - \right.$$

$$\left. - \frac{1}{\sqrt{\pi}} \int_{\frac{1}{2\sqrt{ct}} + \sqrt{s_0 t}}^{\infty} \frac{v e^{-v^2}}{\sqrt{v^2 + 2\sqrt{\frac{s_0}{c}}}} dv \right]$$

(A25)

The integral in the last term is solved by setting

$$\xi^2 = v^2 + 2\sqrt{\frac{s_0}{c}}$$

(A26)

and the final expression is

$$I = \frac{\pi}{2} e^{s_0 t} \left[ 2e^{-\frac{\sqrt{s_0}}{c}} - e^{-\frac{\sqrt{s_0}}{c}} \operatorname{erfc}\left(\frac{1}{2\sqrt{ct}} + \sqrt{s_0 t}\right) - e^{-\frac{\sqrt{s_0}}{c}} \operatorname{erfc}\left(\frac{1}{2\sqrt{ct}} - \sqrt{s_0 t}\right) \right] \quad (\text{A27})$$

Combining Eqs (A16), (A17), (A20) and (A27) the function  $F(a,b,c)$  defined by Eq (A15) can then be written as

$$F(a,b,c) = e^{-at} \left\{ e^{\frac{\sqrt{b-a}}{c}} \operatorname{erfc}\left[\frac{1}{2\sqrt{ct}} + \sqrt{t(b-a)}\right] + e^{-\frac{\sqrt{b-a}}{c}} \operatorname{erfc}\left[\frac{1}{2\sqrt{ct}} - \sqrt{t(b-a)}\right] \right\} \quad (\text{A28})$$

The discharge rate of nuclide No. 3 is finally obtained by the inverse transform of Eq (A10) as

$$N_3(z,t) = \frac{N_3^0}{2} \sqrt{\frac{K_3}{\pi Dt}} e^{-\frac{K_3 z^2}{4Dt}} + A_1 \left[ F\left(a_1, \lambda_1, \frac{D}{K_1 z^2}\right) - F\left(a_1, \lambda_3, \frac{D}{K_3 z^2}\right) \right] + A_2 \left[ F\left(a_2, \lambda_2, \frac{D}{K_2 z^2}\right) - F\left(a_2, \lambda_3, \frac{D}{K_3 z^2}\right) \right] \quad (\text{A29})$$

where  $A_i$  and  $a_i$  are obtained from Eqs (A11) to (A14).

Fig. 1

Relative discharge rates at  $z = 0.5 \text{ km}$ , Impulse release.

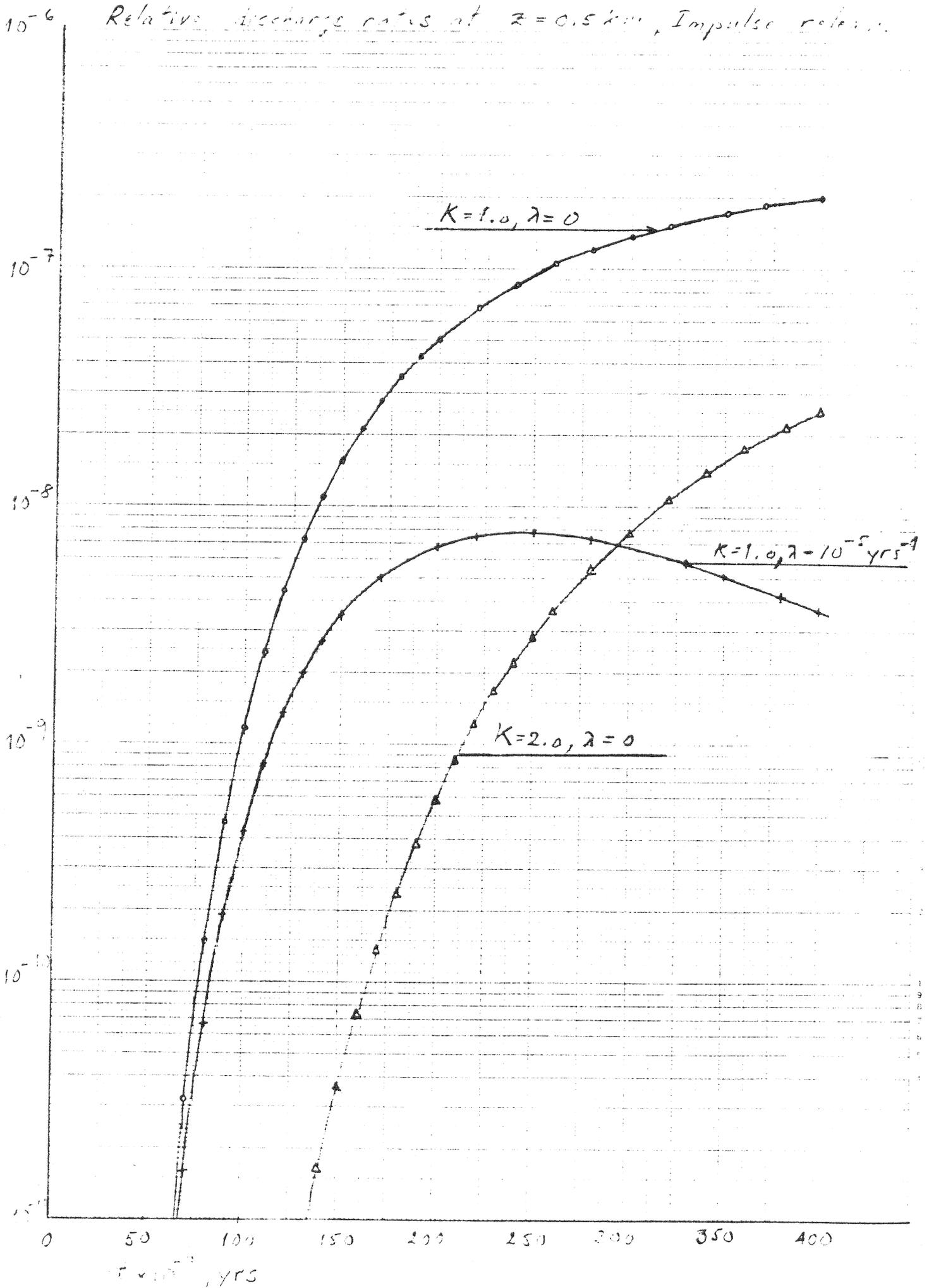
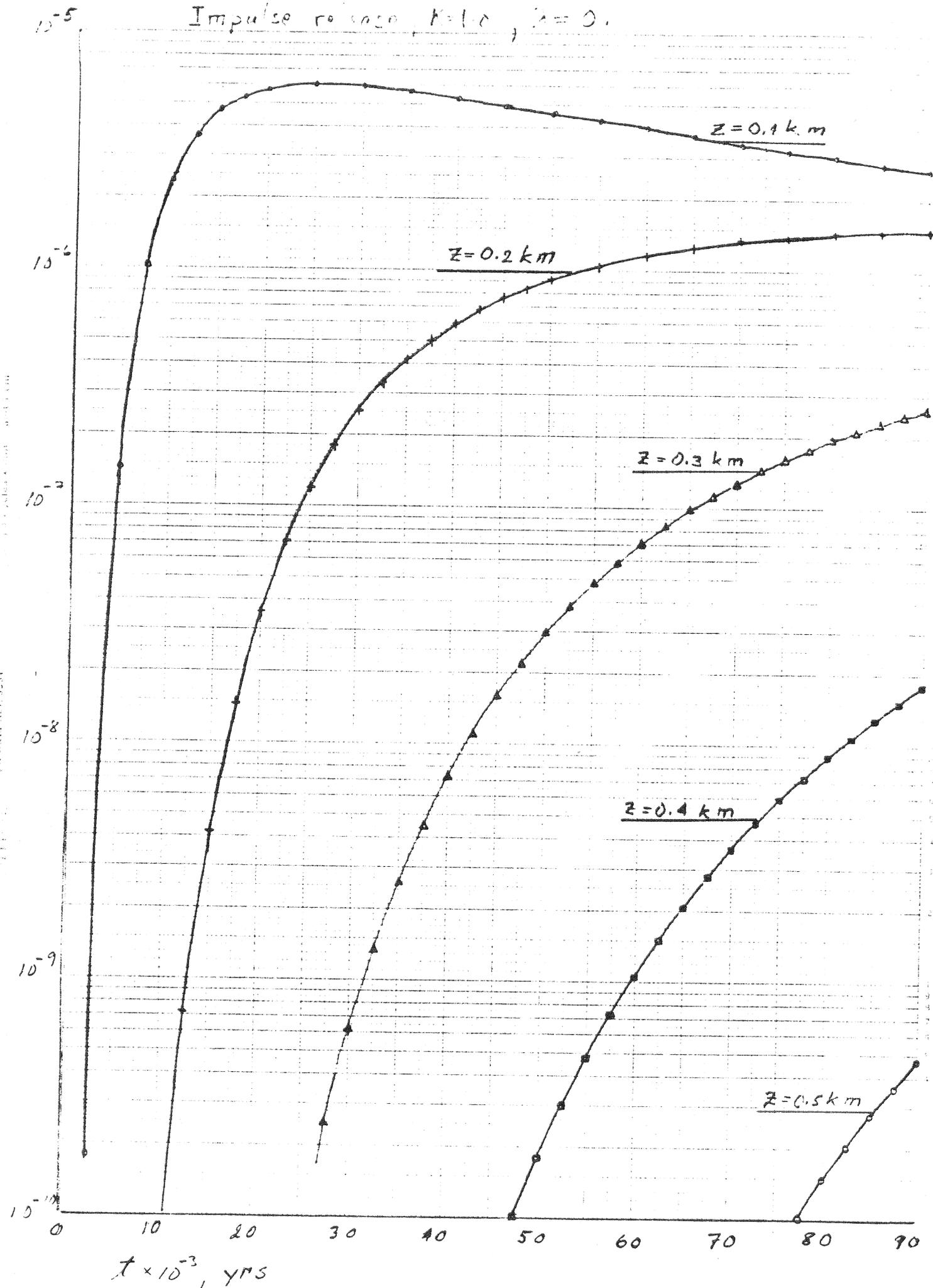


Fig. 2

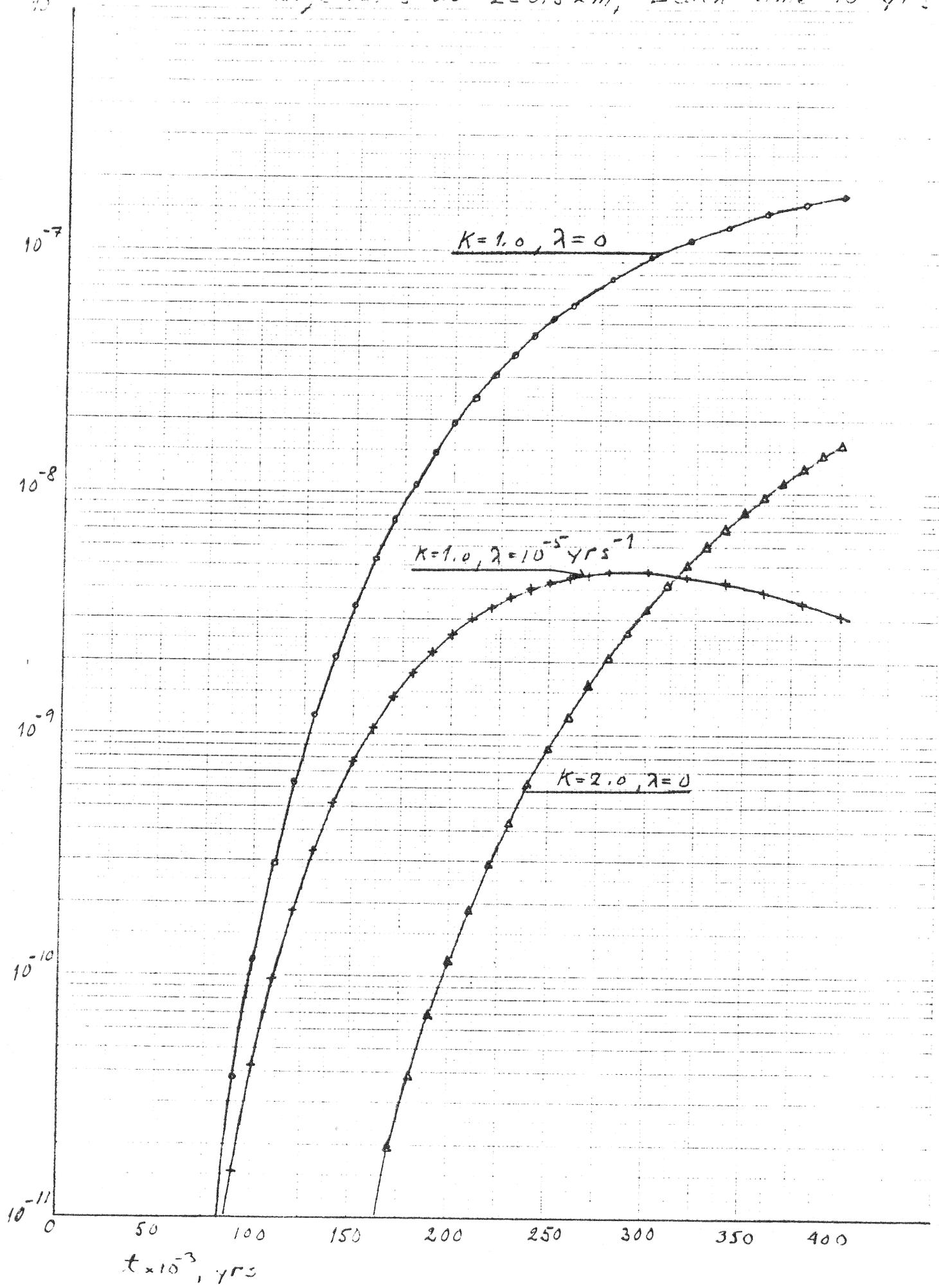
Relative discharge rates at different  $z$ -levels.

Impulse release,  $K=1.0$ ,  $\lambda=0$ .



$\tau = 1, \text{ yrs}$

15% Relative discharge rates at  $z = 0.5 \text{ km}$ , Leach time  $10^5 \text{ yrs}$



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