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## DECONTAMINATION OF RADIOACTIVELY CONTAMINATED WATER USING NATURAL LOCAL MATERIALS

by

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### ABSTRACT

The decontamination of water from radioactive  $^{137}\text{Cs}$ ,  $^{89}\text{Sr}$ ,  $^{144}\text{Ce}$  and their mixture could be achieved using untreated black soil and sand. Black soil gave a higher % of decontamination D than sand when used with  $^{137}\text{Cs}$ ,  $^{89}\text{Sr}$  and the mixture of the three isotopes while sand gave a higher % D than black soil when used with  $^{144}\text{Ce}$ . The mechanisms of fixation of radionuclides on the decontaminant have been explained by the ion-exchange, physical adsorption and the gel filtration mechanisms.

### 1. INTRODUCTION

Supply of troops by fresh water following a nuclear attack represents a serious problem. It is known that synthetic adsorbents and ion-exchange resins could be used to remove radionuclides from solutions. However, the supply with those materials and the conditions of their use may represent a problem during the combat action. The present work illustrates the use of available local material such as sand and black soil in decontaminating water from the most dangerous radioactive isotopes in the fission products mixture :  $^{137}\text{Cs}$  ( I ),  $^{89}\text{Sr}$  ( II ) and  $^{144}\text{Ce}$  ( III ). This also represents a method for the treatment of low level radioactive waste [1]

### 2. EXPERIMENTAL

Materials ; Radioactive nuclides used were  $^{137}\text{Cs}$  ( I ),  $^{89}\text{Sr}$  ( II ) and  $^{144}\text{Ce}$  ( III ). The counting rate was determined using counter NZQ 612 Tesla; and the sieving machine "Stavebni stroje n.p. Zlioin" was used in preparing the different sizes of the materials.

#### Procedure :

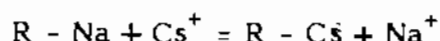
Four solutions of common tap water ( PH = 7.5 ) were contaminated with Sr, Cs, Ce and their mixture. The initial counting rate  $I_0$  was in the range 2000 c.p.m/ml. 20 ml of the contaminated water were added to 5 g of the decontaminating material in a polyethylene bottle. The system was shaken for one minute and the residual counting rate  $I_1$  of water was counted each 5 minutes interval ( starting from the addition ) for 120 minutes. The percentage of decontamination D was calculated as follows :

$$D = \frac{I_o - I_r}{I_o} \times 100 \quad (\%)$$

### 3. RESULTS AND DISCUSSION

Preliminary experiments showed that the percentage of decontamination reached a constant value when 4-5 g of the decontaminating materials were used. Thus, 5 g of each material were used.

Fig. 1, 2 and 4 show that black soil gave a higher % D (>80%) than sand for  $^{137}\text{Cs}$ ,  $^{89}\text{Sr}$  and for the mixture of the three isotopes used. On the other hand Fig. 3 indicates the higher % D of sand than black soil for  $^{144}\text{Ce}$ . To explain this, the state of the radionuclide in solution, the characteristics of the surface and the mechanism of fixation of the radionuclide on the decontaminant should be discussed [2]. Both  $\text{Cs}^+$  and  $\text{Sr}^{2+}$  exist mainly in the form of ions till  $\text{PH} = 11$  [3] which explains their mechanism of fixation on black soil and sand. Black soil used in this study has been found to be an aluminum silicate containing mainly  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and some  $\text{Fe}^{3+}$  [3] These ions are present mainly to compensate the net negative charge on the surface caused by the isomorphous replacement of  $\text{Mg}^{2+}$  for  $\text{Al}^{3+}$  in the octahedral sheets and  $\text{Al}^{3+}$  for  $\text{Si}^{4+}$  in the tetrahedral sheets in the aluminosilicate structure [4,8]. Thus, the fixation of both  $\text{Cs}^+$  and  $\text{Sr}^{2+}$  on black soil could be explained by the ion exchange between  $\text{Cs}^+$  in solution and  $\text{Na}^+$  on the surface as well as  $\text{Sr}^{2+}$  with  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ . The ion exchange mechanism could be schematically represented as such :



In case of  $^{144}\text{Ce}$ , it has been shown that [3,5]  $^{144}\text{Ce}$  exists in the form of positively charged hydrolysed species  $\text{Ce}(\text{OH})_2^+$ ,  $\text{Ce}(\text{OH})_3^+$  between  $\text{PH} 4-7$  which can not be easily exchanged with the cations on the surface due to their larger size.

On the other hand at  $\text{PH}$  higher than 7, as in our case,  $^{144}\text{Ce}$  bears a negative charge [5] ( $\text{Ce}(\text{OH})_4^-$ , ...) while the charge on the clay surface is also negative. The repulsion between  $\text{Ce}$  and the clay explains the lower % D in case of  $\text{Ce}$  than with  $\text{Cs}$  and  $\text{Sr}$ . Considering sand which is mainly silica ( $\text{SiO}_2$ ), it is generally accepted that silica surface bears a negative charge above its isoelectric point at  $\text{PH} = 2$  [6]. Thus, the fixation of  $\text{Cs}^+$ ,  $\text{Sr}^{2+}$  and positively charged species of  $\text{Ce}$  on sand could be related to the electrostatic attraction between the negative adsorption on the surface.

Fig. (5) shows that smaller sizes of sand grains gave a higher % D than the larger ones which indicates the increase of the physical adsorption due to the increase in the surface area.

If cation exchange and physical adsorption are the only mechanisms responsible for fixation of radioelements on the contaminant surface then black soil should show a higher % D than sand with  $^{144}\text{Ce}$  as with  $^{137}\text{Cs}$  and  $^{89}\text{Sr}$ . but this is not the case. Fig. (3) shows that sand gave a higher % D for  $\text{Ce}$  than black soil. Thus, it is reasonable to think of another mechanism such as :

Gel filtration. on the voids of the silica structure and in this case the bigger the size of the radioelement species the higher will be the gel filtration. This may explain the reason of higher % D with sand than with black soil in case of  $^{144}\text{Ce}$  .

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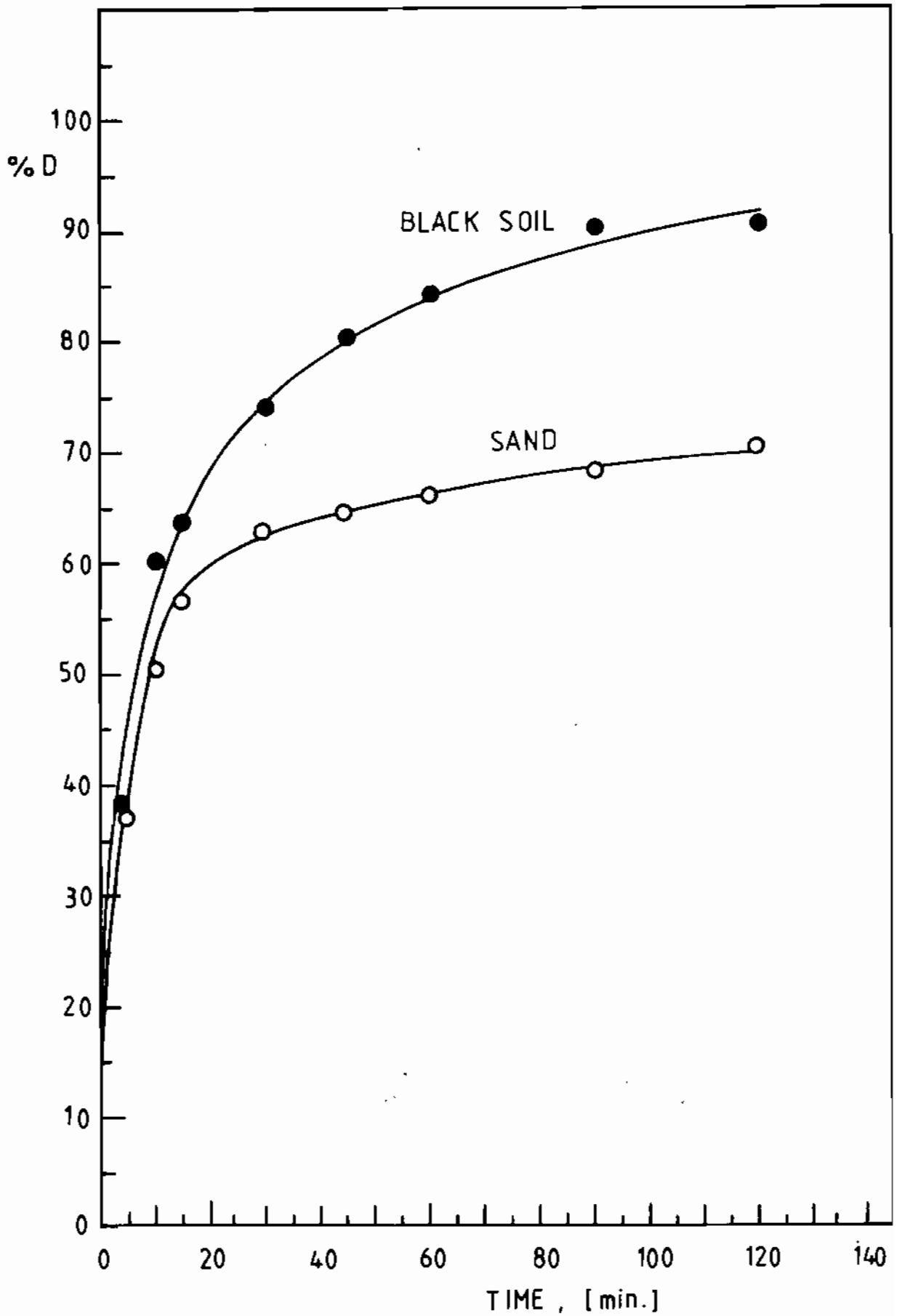
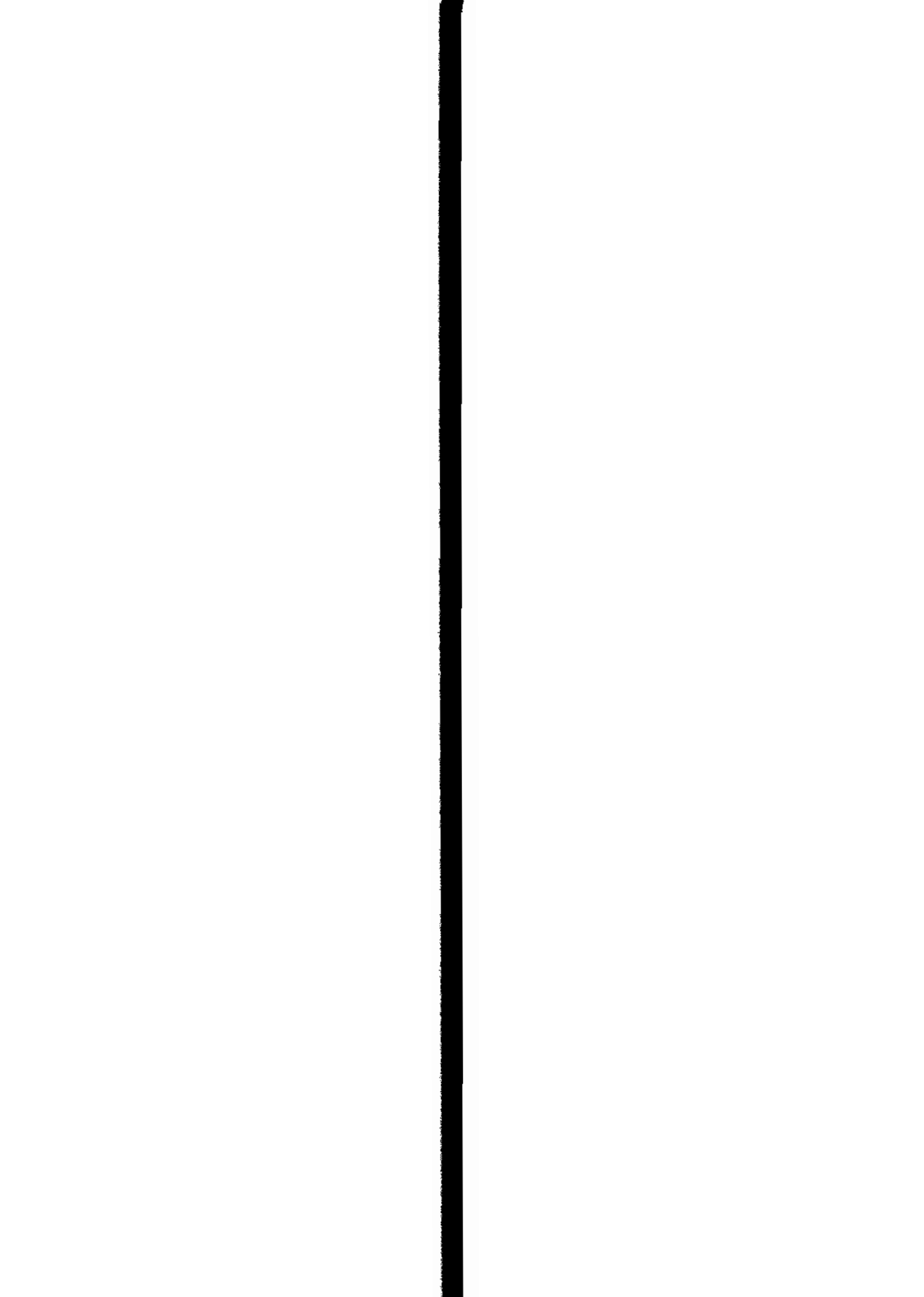


Fig. (1): Dependence of %D on time for  $^{137}\text{Cs}$ .



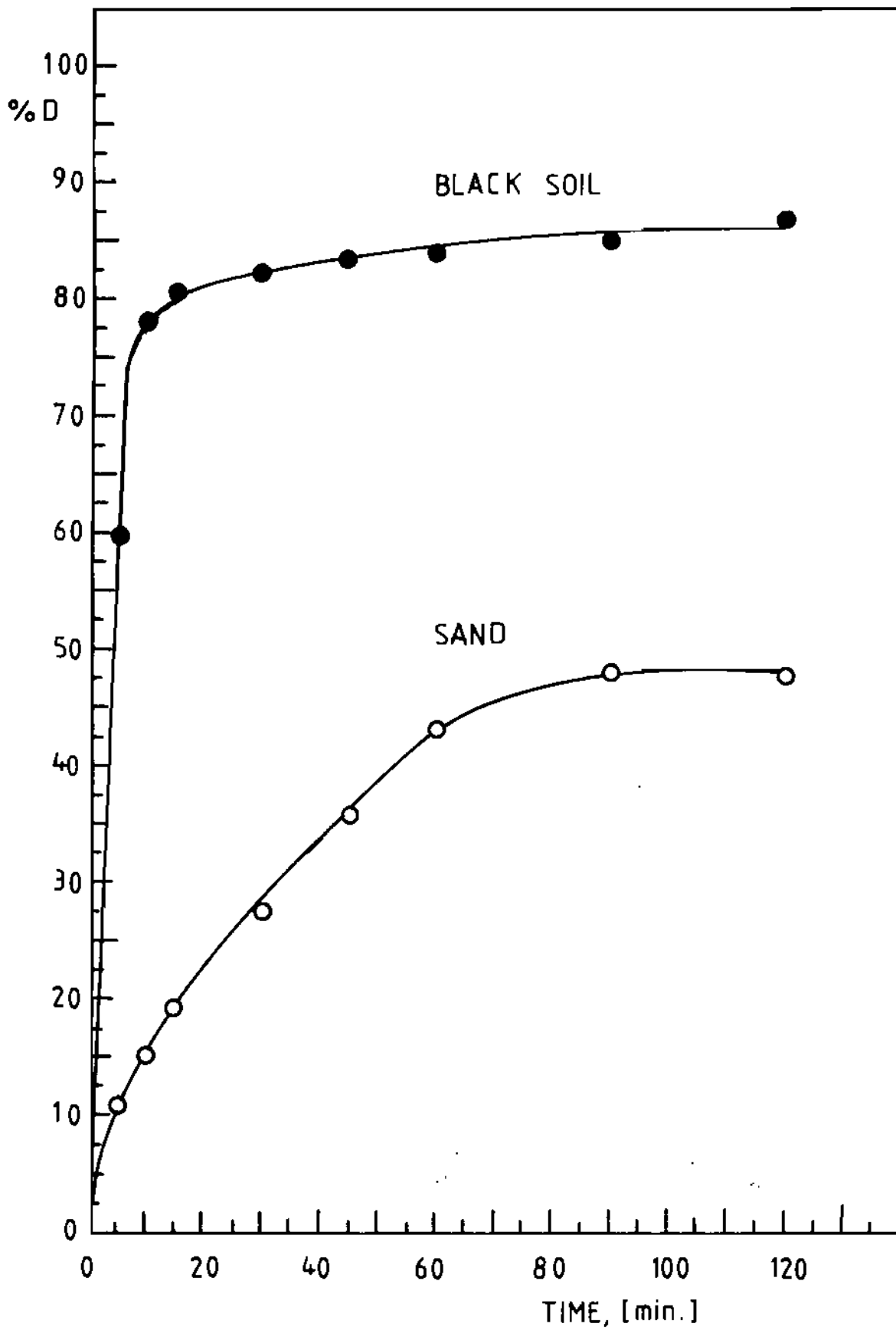


Fig.(2): Dependence of %D on time for  $^{89}\text{Sr}$ .

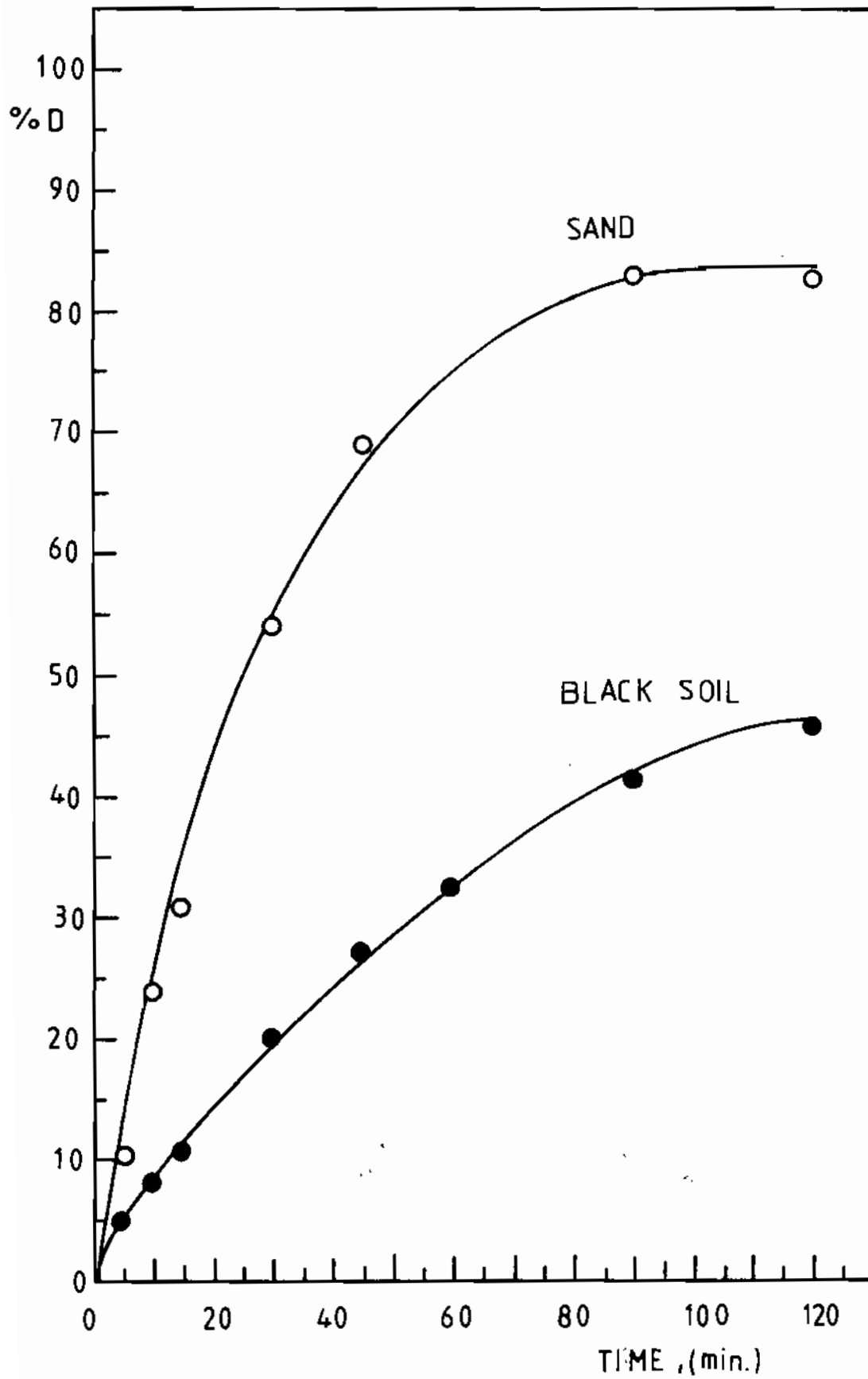


Fig.(3) : Dependence of %D on time for  $^{144}\text{Ce}$ .



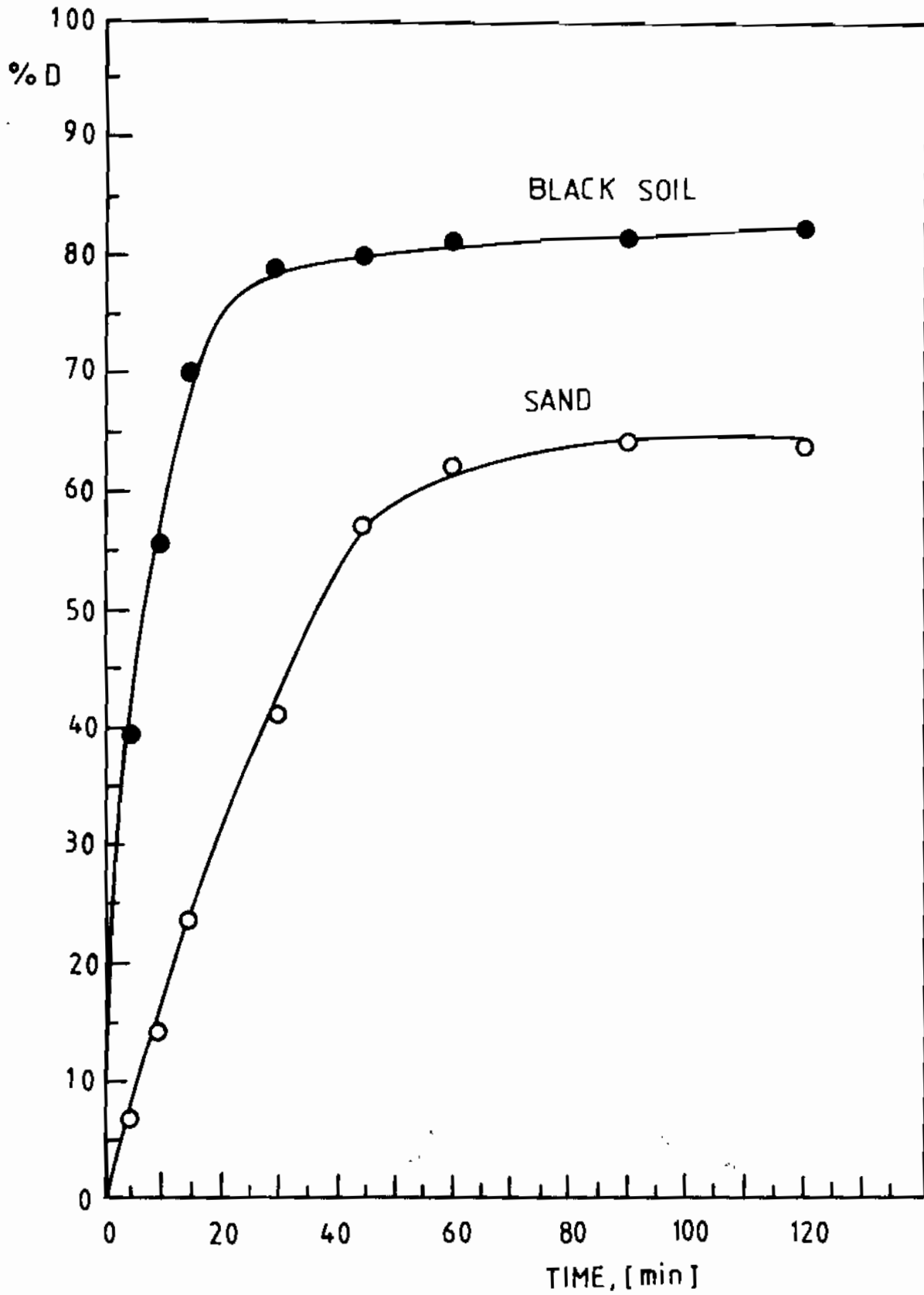


Fig.(4): Dependence of % D on time for  $^{137}\text{Cs}$ ,  $^{89}\text{Sr}$  &  $^{144}\text{Ce}$  mixture.

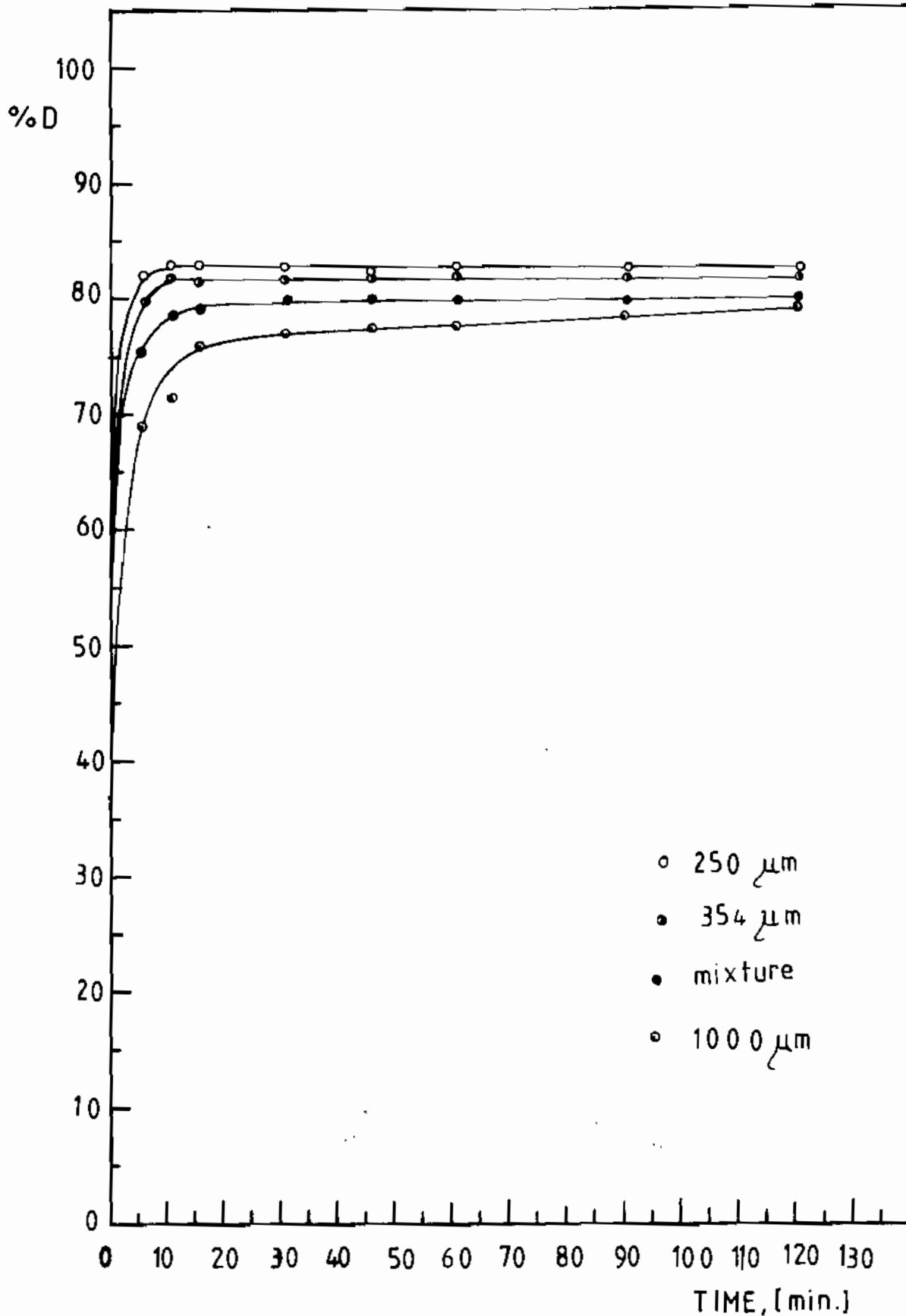


Fig.(5): Effect of grain size of sand on % D for  $^{137}\text{Cs}$ ,  $^{89}\text{Sr}$  &  $^{144}\text{Ce}$  mixture.