TRANSFER OF FISSILE MATERIAL THROUGH SHIELDING COATINGS IN EMERGENCY HEATING OF HTGR COATED PARTICLES

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ABSTRACT

The measurement results of leakage dynamics of fissile material from the coated particles within a temperature range of 1200 ÷ 2000°C are given. The methods of carrying out the experiments are briefly described. The relation of the leakage rate of uraniun-235 from CP with the pyrocarbonic coatings has been obtained.

The measurement of the fissile material (FM) leakage from the HTGR CP at temperatures of exceeding 1000°C is interesting both for predicting the aftereffects of the fuel emergency overheating and the evaluation of the matrix graphite contamination in the production of ball CPEL. The results of the investigations (1,2) show that at temperatures of exceeding 1500°C a considerable transfer of uranium is observed in the carbonic CP coatings. At the same time no data on the uranium yield from CP have been published. The aim of the present work is to investigate the FM leakage from BiSo CP within a temperature range of 1200 ÷ 2000°C.

The experiments have beeen carried out at the plant including the vacuum chamber with a device for heating CP and sampling the fissile material. The FM settling from the molecular flow leaving the sample takes place on the cold changeable collectors during the pre-determined time of exposure. The uranium amount on the collectors is measured by the radiographic method of the fission fragment tracks after which the FM number is calculated which has left the CP for the time of exposure. The sample temperature is measured by a pyrometer. The pressure during the experiment does not exceed IC^{-6} torr.

Fig.I gives the graphs of the relations of the uranium-235 leakage rates and time at a temperature of I900°C from the micro CP of Type BiSC from HTI and LTI layers of dense pyrocarbons. The characteristic peculiarities of the presented results:

- fast ($<$ IC minutes)reach of the stationary leakage rate of the both CP types;
- a considerable difference in the equilibrium rates of leakage for the two CP types.

Fig.2 gives the graphs of the relations of the FM leakage from CP and annealing time. It also shows the time relations of the uranium-235 in the coating and core of the both CP types. A rather surprising fact can be considered that the FM amount having come out of CP with the HTI coating in 3C minutes exceeds one order the FM content in the coating and by IC hours of annealing makes up already procents of the initial FM amount in CP. The FM leakage from CP with the LTI pyrocarbon by IO hours makes up IC^{-3} of the initial content in CP that is approximately equal to the FM content in the coating to the same time.

For cheching up a direct measurement of FM loss from CP for the annealing time taking into consideration a change in the intensiti of gamma radiation with an energy of I85.7 keV has been carried out. The results of the gamma spectrometric measurements match the results of measurements of the F flow from the sample.

Fig.3 gives the results of the investigation (3) in the simultaneous measurement of the cesium flow through the graphite memebrane and Cs amount accumulated by the membrane. The construction of the curves in Figure 3 qualitatively coincides with the analogous curves for uranium in Figures I and 2 , namely:

- quick establishment of the stationary flow in the membrane outlet for Cs and outlet of the PyC coating for uranium;
- continuing of the Cs accumulation in the memebrane and uranium in the coating after establishment of the stationary flow in the outlet.

It can be supposed that the CS transfer mechanism as well as uranium in the carbonic materials is just the same. In this case, on the one hand, the uranium transfer can be described within the model limits suggested in (4) for describing the results of the investigation (3), on the other hand, the investigation of the uranium transfer regularities can be considered as the modelling of the cesium transfer in the CP coatings. In favour of the assumption of the similar mechanism of the cesium and uranium transfer in the CP coatings it is possible to also give the other experimental results:

- characteristic peculiarities of the Cs and U propagation in BiSC particles are equal, Fig.4;
- for Cs and U a comparatively high mobility is characteris-

tic in the pyrocarbonic layers of the coatings and for Cs and U layer SiC serves as the main barrier.

Fig.5 presents the time relations of the uranium-235 from CP with the dense PyC coating of type HTI in the temperature range of 1200 ÷ 1900°C. Fig.6 shows the relation of the meam leakage rate from the same samples and the reverse temperature which is well described by Equation $\bar{R}u = R_o \exp(-Q/RT)$ with an activation energy of $Q = 76 \pm 6$ kCal/mol.

By the the results of the work it is possible to formulate the following conclusions:

- there exists a considerable leakage of the fissile material from BiSC CP at a temperature 1200°C and above. At a temperature of 1900°C BiSC CP with the HTI coating loses for several hours some percents of the initial amoumt of the fissile material;
- the characteristic peculiarities of the leakage dynamics and accumulation in diffusion of uranium and cesium atoms through the CP coatings rather resemble which permits to suppose the similar transfer mechanism of these nuclides and a possibility of using the results of investigation of the uranium behavour for describing the cesium migration;
- mean rate of the uranium-235 leakage from BiSC CP can be described by relation $\bar{R}u = R_o \exp(-Q/RT)$ with an activation energy of $Q = 76 \pm 6$ kCal/mol in the temperature range of 1200 ÷ 1900°C.

R E F E R E N C E

I. R.W.Daton, J.N.Cxley, C.W.Townley. Ceramic Coated Particle Fuels. Journ.of Nucl.Mater., I964, V.II, No.I, pp.I-34.

2. A.N.Gudkov, A.A.Kozap, A.D.Kurepin. Peculiarities of Profile Formation of Fissile Material Concentration in HTGR Coated Particles. Voprocy atomtoy nauki n tekhniki. ser.Atomto-vodorodnaya energetika i tekhnologiya. Vyp.I, I988, str.70-7I.(Russian)

3. A.B.Riedinger, C.E.Milstead, L.R.Zumwalt. Proc.Fifth Carbon Conf. I963, V.II, p.405.

4. D.Chandra, J.H.Norman. Diffusion of Cesium through Graphite. Journ.of Nucl.Mater., I976, V.2, No.2,pp.293-3IO.

5. Zoller P. Das Transportverhalten der Spaltproducte Cesium und Stroncium in beschichteten Brennstoffteilchen fur Hochtemperaturreaktoren unter Bestrahlungen. Jul-I324, Julich, I976.

CAPTIONS TC FIGURES

Fig.I. Uranium leakage rate from BiSC CP, temperature I900°C:
I - LTI PyC, 2 - HTI PyC.

Fig.2. Uranium leakage dynamics from BiSC CP and uranium content in PyC, temperature I900°C:
I - LTI, leakage from CP; 2 - LTI, content in PyC; 3 - HTI, leakage from CP; 4 - HTI, content in PyC; 5 - LTI, HTI - content in kernel

Fig.3. Cs leakage and accumulation dynamics in its diffusion through graphite memebrane, (3):
I - leakage rate; 2 - leakage; 3 - content in memebrane.

Fig.4. Uranium and Cs concentration profiles in BiSC CP:
I - uranium; 2 - Cs.(5)

Fig.5. Uranium-235 leakage from BiSC CP with dense PyC layer of HTI type.

Fig.6. Relation of uranium-235 mean leakage rate and reverse temperature.

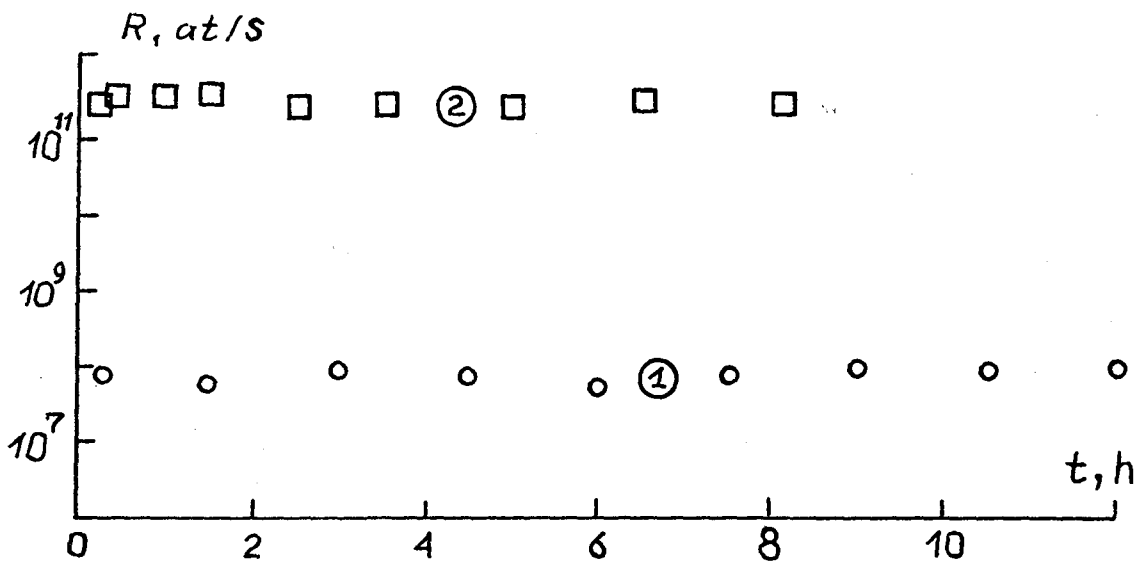

Fig. 1

A–5

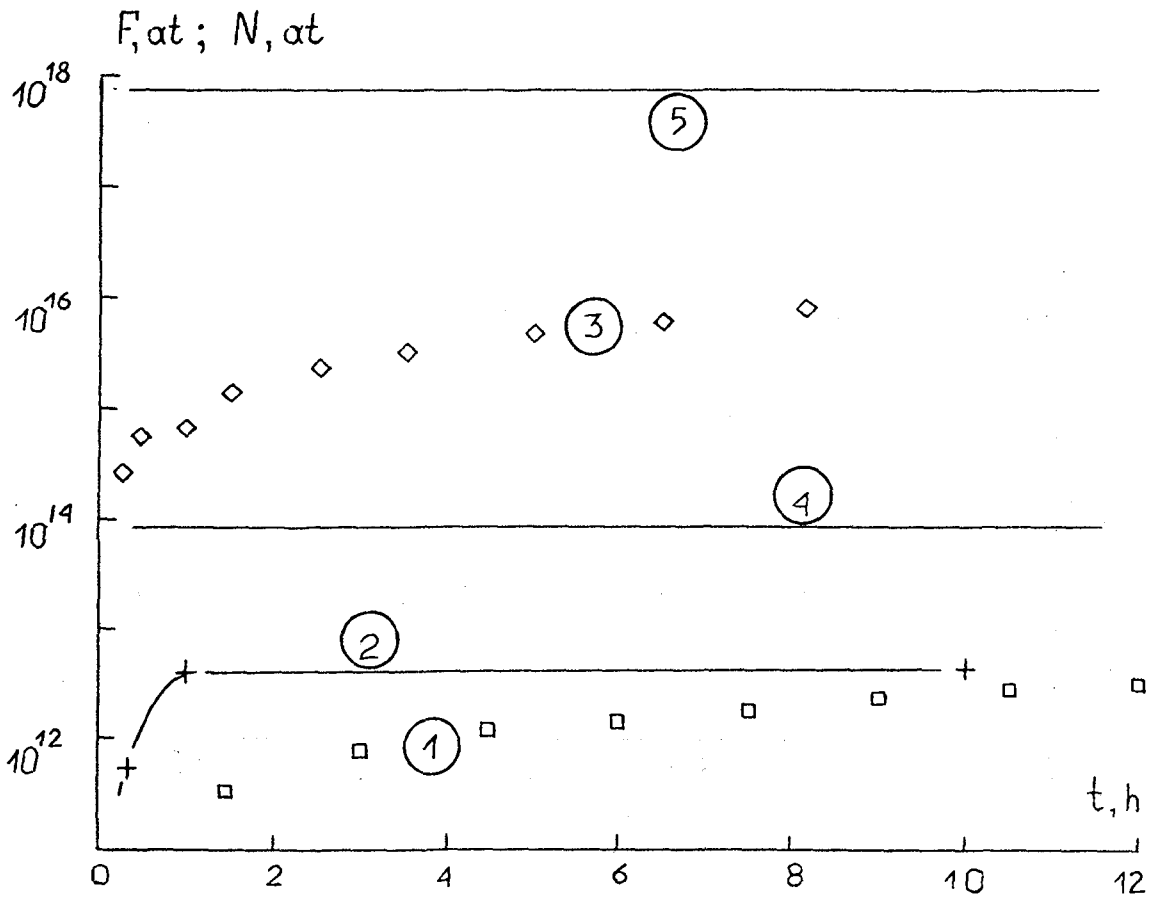

Fig. 2

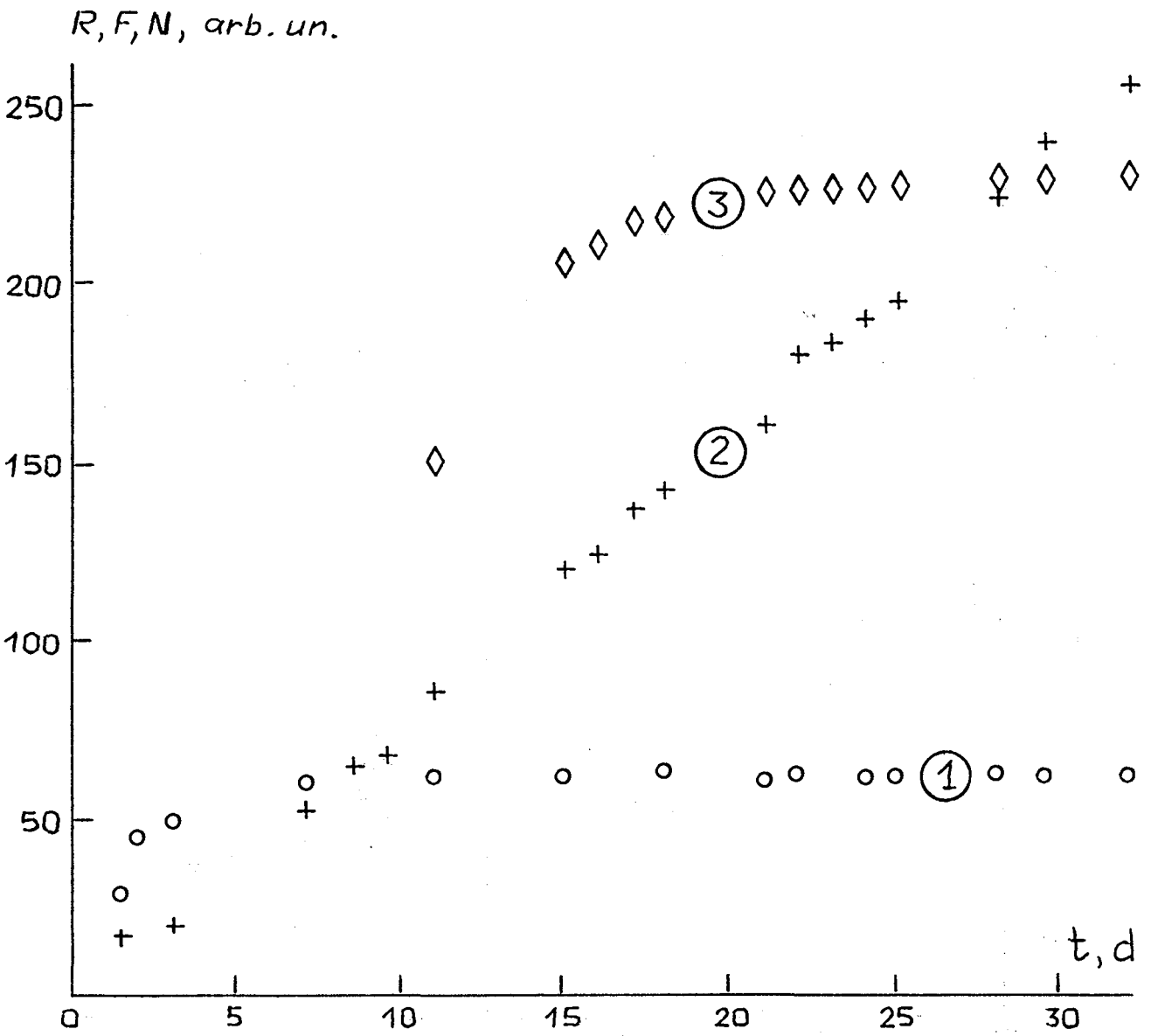

Fig.3

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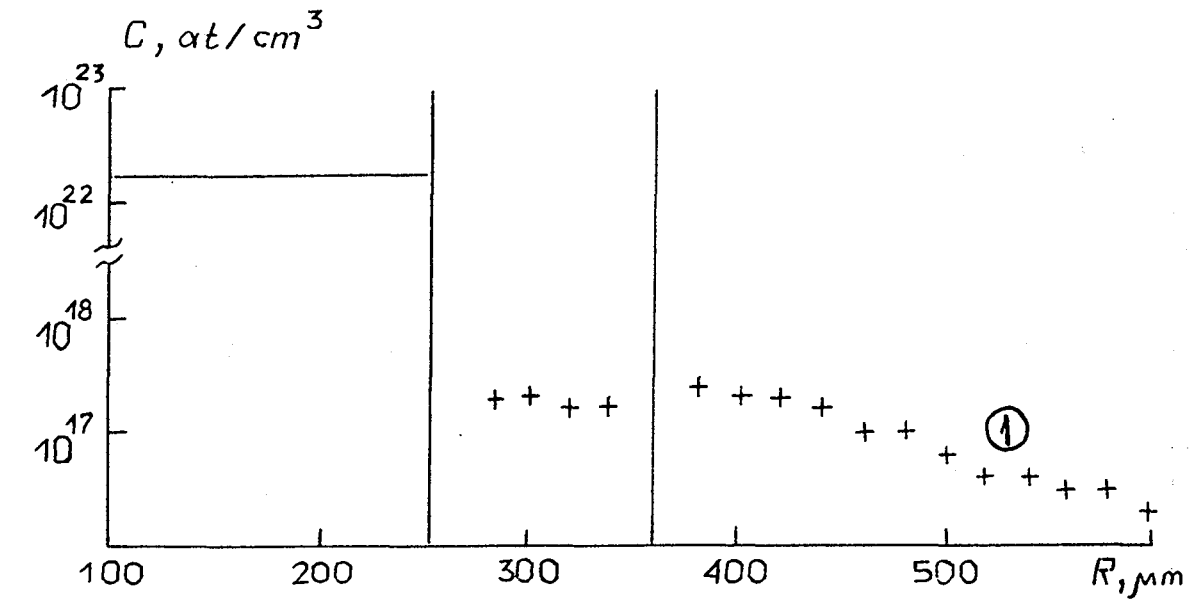
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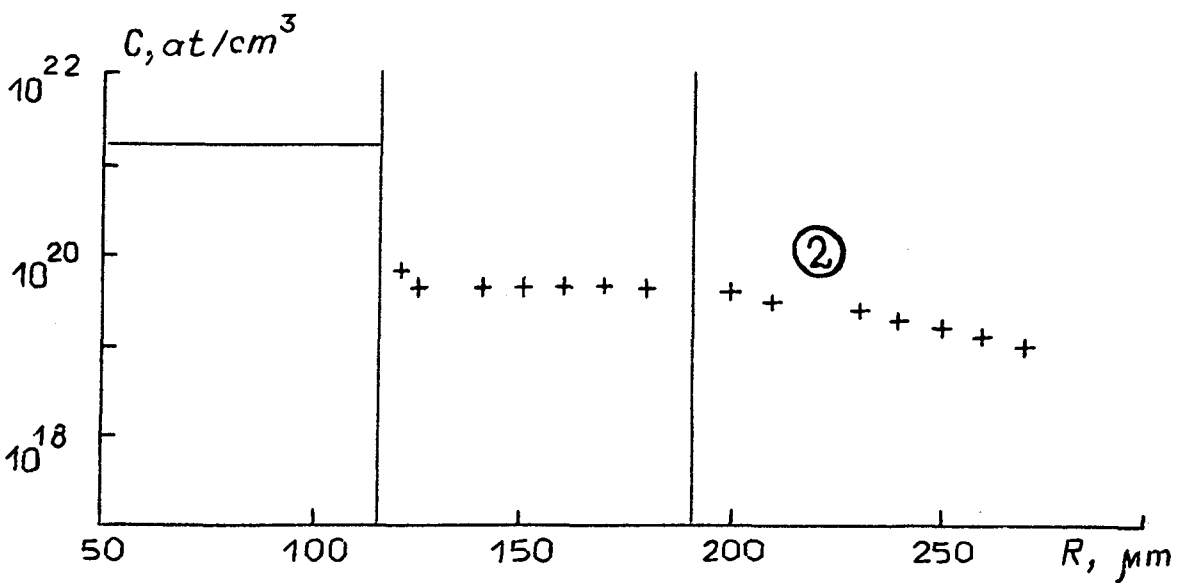
10²²
10¹⁸
10¹⁷
100
200
300
400
500
R, мм
①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10²⁰
10¹⁸
50
100
150
200
250
R, мм
②

Fig. 4

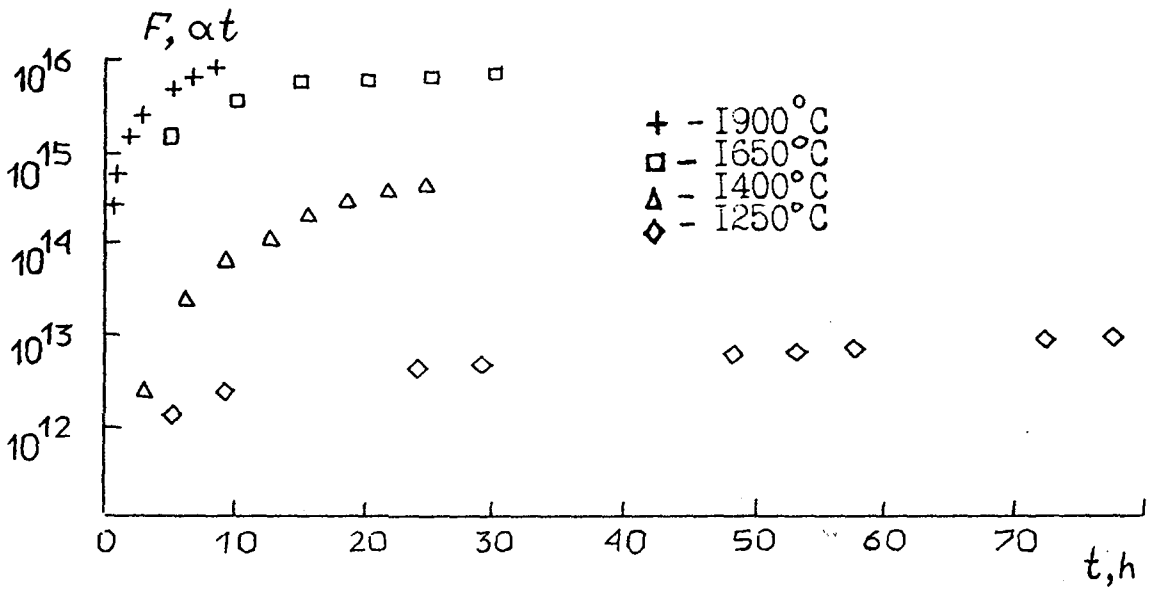
F, at
10^{16}
10^{15}
10^{14}
10^{13}
10^{12}
+ - I900°C
□ - I650°C
△ - I400°C
◇ - I250°C
0
10
20
30
40
50
60
70
t,h

Fig.5

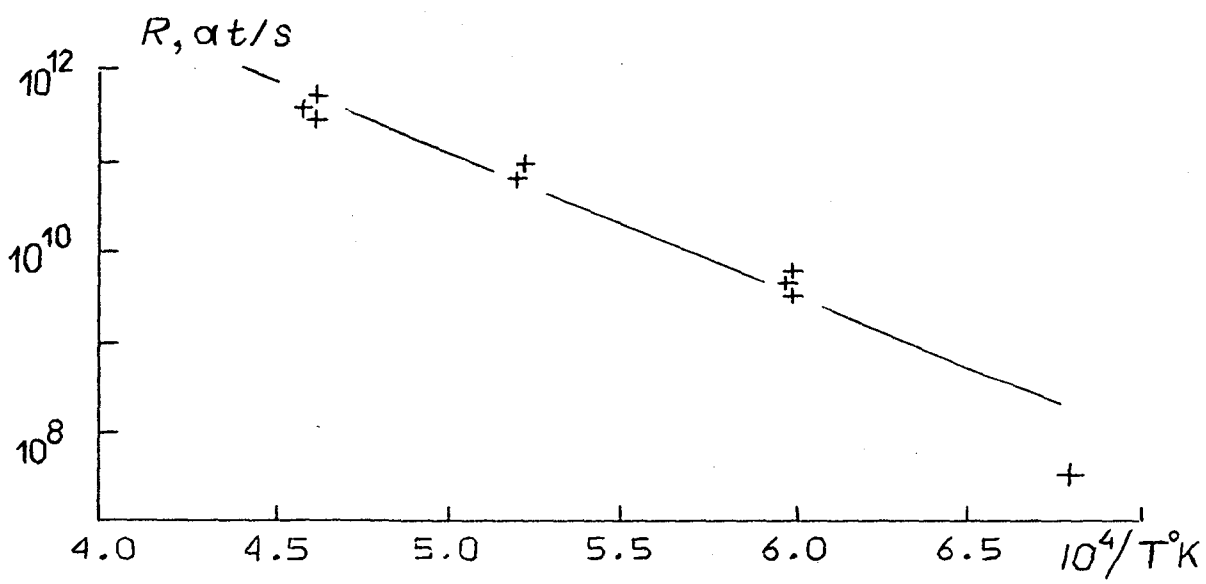
R, at/s
10^{12}
10^{10}
10^{8}
4.0
4.5
5.0
5.5
6.0
6.5
$10^4/T°K$

Fig.6