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PRELIMINARY INTERPRETATION OF THE LIBRETTO-2 EXPERIMENTAL RESULTS IN TERMS OF TRITIUM PERMEATION BARRIER EFFICIENCY

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PRELIMINARY INTERPRETATION OF THE LIBRETTO-2 EXPERIMENTAL RESULTS IN TERMS OF TRITIUM PERMEATION BARRIER EFFICIENCY

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ABSTRACT

Within the framework of a European Community programme aiming at the development of a water-cooled lithium-lead (Pb17Li) blanket concept for a demonstration fusion reactor, important experimental works are devoted to the development of efficient tritium permeation barriers. Among them, in-pile tests (LIBRETTO experiments) with closed capsules containing stagnant Pb17Li are intended to assess under irradiation the efficiency of these barriers.

In this paper, a method for interpreting LIBRETTO data is presented, which relies on the use of a 2-dimensional finite element simulation model exploiting the analogy between tritium migration and heat conduction. Preliminary results obtained with this model are also discussed, which seem to indicate that, under conditions comparable with the experiment, the permeation barrier tested in LIBRETTO-2 would reduce tritium permeation by a factor of about 80.

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1. INTRODUCTION

The control of tritium permeation through the cooling tubes to the water coolant is a key feasibility issue for water-cooled lithium-lead blankets (Malang [1]). It requires the development of efficient permeation barriers which have to be qualified under representative irradiation conditions. The LIBRETTO 2 irradiation experiment (Conrad [2]) is the very first step of this development and qualification process.

Up to now, the exploitation of the results of this experiment has been limited to a **relative** comparison of the efficiency of the tested permeation barriers. This kind of **qualitative** interpretation is very useful for selecting the type of barrier to be further developed. However, only a **quantitative** interpretation, in **terms of permeation reduction coefficient of the barrier**, will permit to progress towards the demonstration that tritium permeation control is feasible.

In order to perform this quantitative interpretation, a computational model has been developed, which simulates the transient migration of tritium in LIBRETTO-type Pb17Li capsules.

This paper describes this model and the associated method proposed for deriving a permeation reduction coefficient from the comparison of the numerical simulation results with the experimental measurements. Some preliminary results are also presented and discussed.

2. THE LIBRETTO 2 EXPERIMENT

The LIBRETTO 2 experiment, jointly carried out by JRC Ispra, CEA, JRC Petten and performed by JRC/Petten, consisted in irradiating, in the HFR reactor, 4 independently controlled, triple contained and closed capsules (*Figure 1*) made of steel and containing Pb17Li in liquid form under an helium ceiling, and monitoring the amount of the tritium generated within Pb17Li which permeates through the capsule walls by measuring the activity of the helium flow sweeping the outside of the capsule (**Conrad [2]**).

The various capsules tested in LIBRETTO 2 differ by the type of steel, the volume of the helium plenum, the presence or not of a permeation barrier.

Thus, the LIBRETTO 2 experiment constitutes the first attempt to investigate under irradiation phenomena of diffusion in Pb17Li and permeation through steel of tritium generated within Pb17Li, and the influence of permeation barriers.

The only known tentative interpretation of this experiment was made in terms of tritium residence time in the capsule: the use of permeation barriers was shown to increase this residence time by a factor 4.5 (**Conrad [2]**).

This way to quantify the efficiency of the tested barriers is of little usefulness for the blanket design and for the designer of the water detritiation unit ; indeed both want to know the coefficient by which a given permeation barrier will reduce the amount of tritium permeating through a water tube of given thickness. The development of a computer tool and of its associated method for interpreting LIBRETTO results in terms of this so called "permeation reduction coefficient" was the purpose of the study reported hereafter.

3. A COMPUTER MODEL SIMULATING TRITIUM TRANSPORT IN LIBRETTO TYPE CAPSULES

It is generally admitted that the migration of tritium within solid or stagnant fluids with negligible temperature gradients follows the classical diffusion equations, like heat transfer.

Therefore, the versatility of the 2D finite elements heat transfer code DELFINE [3] was taken benefit of to exploit this analogy between tritium transport and heat conduction and develop a numerical model simulating, under transient conditions, the migration of tritium in a LIBRETTO-type capsule.

3.1 The Analogy

According to the following notation: **C** the tritium concentration, **D** the tritium diffusion coefficient, **K** the tritium solubility coefficient, **P**: the tritium partial pressure, **Qr** the recombination rate constant, **T** the temperature, **R** the perfect gas constant, **H** the heat exchange coefficient, ϕ the tritium permeation flux, and subscripts s, l, h and g for respectively steel, lithium-lead, helium plenum, and sweep gas; the analogy considered for a capsule without permeation barriers is as follows :

.**for steel and Pb17Li**, the ratio **C/K** (equal to $P^{1/2}$) is analogous to a temperature, the product **D * K** to a thermal conductivity, and **K** to a volumetric specific heat

.**for helium**, the square root of **P** is analogous to a temperature, while the product $2 R T_h P_h^{1/2}$ is analogous to a volumetric specific heat

.and **at the capsule steel wall/ sweep gas interface**, if a recombination law $\phi = Qr(P_s - P_g)$ is assumed, the product $Qr(P_s^{1/2} + P_g^{1/2})$ is analogous with a heat exchange coefficient a recombination law $= Qr(P_s - P_h)$ is adopted, where is the permeation flux

Under these conditions, the continuity of both the tritium partial pressure and the tritium flux is respected.

3.2 Limitations of this analogy

The main limits of this analogy are the following :

- the limiting step in the transport of tritium from Pb17LI to the helium plenum is assumed to be diffusion within Pb17Li. Actually, tritium generation by ^{6}Li being accompanied with helium production, rising helium bubbles could significantly participate in tritium transport (as well as natural convection). This limitation can be passed around by artificially increasing D_l in the axial direction.

- the dissociation phenomenon $T_2 \text{-->} 2\,T_{ads}$ at the helium plenum/steel interface is not explicitely taken into account. This model limitation could however be easily compensated for by taking Ks as the ratio *P*s/Ds (*P*s: permeability) instead of the solubility coefficient.

- the same holds for the Pb17Li/steel interface as well, and the same artifice could also be used. It is not clear whether such a recombination/dissociation occurs at this interface as tritium is generated within Pb17Li directly under atomic (and not molecular) form. However, helium bubbles sticking at the capsule wall (a phenomenon which was observed during the post-irradiation examinations of LIBRETTO) could force tritium to recombine before it reaches the capsule walls.

4. FIRST RESULTS OBTAINED WITH THE SIMULATION MODEL

The above described model was run to tentatively simulate the tritium migration behaviour of the LIBRETTO capsule 6 (a case with stainless steel 316 L walls, a large helium plenum, and no permeation barrier) during the test run where it was maintained at nearly constant temperature (300°C) and tritium generation rate starting from a state at zero tritium concentration.

A 2-dimensional (R-Z) finite element mesh comprising 300 elements, including 200 for the steel walls, was used to describe the capsule. Properties data adopted for stainless steel 316 L and lithium-lead are obtained from **Forcey [4]** and **Reiter [5]** respectively. The value of the recombination rate constant Qr is calculated according to the method proposed by **Baskes [6]**, assuming a "sticking factor" of 10^{-3}.

The comparison of the permeation rate history observed during the experiment with the ones resulting of some typical simulations is illustrated in *Figure 2*.

Although the two curves exhibit nearly the same initial slope, the model appears to predict a time to reach steady state much shorter than the one observed, when reference material properties data are used.

If the value of the tritium diffusion coefficient of Pb17Li along the capsule axis is increased by a factor of 20 ($D_l^y = 20\ D_l$) to tentatively simulate the effect of rising helium bubbles or natural convection, a closer fit with the experiment is obtained in both the initial and final phases of the transient. A larger increase of D_l^y leads to a strong divergence in the final phase, and loosen the agreement on the initial slope.

If on the contrary the "sticking factor" (which caracterizes of the state of cleanliness of the capsule wall) is reduced from 10^{-3} down to $2\ 10^{-7}$ (a somewhat unrealistic value), a better fit throughout the transient is obtained, but the initial experimental slope is not followed.

As the value of D_l^y essentially influences the time to reach the steady state tritium inventory in the helium plenum, the test run on capsule 7 (similar to capsule 6 but for the volume of the helium plenum, which is one eighth of the one of capsule 6) was also simulated. Differences between simulation and experiment prove to be quite similar.

These very first and partial results are presented to give an idea of the capabilities of the proposed simulation model, and of the DELFINE code in which it is implemented. Although quite encouraging, they demonstrate that more work and sensitivity studies are necessary not only to discriminate between the various possible boundary conditions/ interface laws, but also to validate the material properties data. In this respect, beyond further analyses (especially of temperature transient tests) and model improvements, experimental data on the out-of-pile permeability of steels in presence of Pb17Li are clearly needed to progress in the interpretation of such in-pile experiment as LIBRETTO.

In this matter of interpretation, one of the main interests of the proposed model is that it should permit to determine the value of the permeation reduction coefficient of the tested barriers. The next section is devoted to a presentation of the proposed interpretation method.

5. A TENTATIVE METHOD FOR INTERPRETING THE PERMEATION BARRIER TESTS IN TERMS OF PERMEATION REDUCTION COEFFICIENT

5.1 Case with barrier on the upstream side

If one assumes that the permeation inhibition effect of the barrier is the one illustrated in *Figure 3*, that the barrier either (a) exhibits a very low diffusion coefficient, or (b) works by preventing the dissociation $T_{2gas} \text{-->} 2T_{ads}$, then the case of a capsule equipped with an upstream barrier can be modelized by artificially reducing appropriately the tritium solubility coefficient of steel.

Thus the method proposed for deriving the value of the permeation reduction coefficient consists of four steps :

Step 1 : confirmation of D_l^y values and interface laws by fitting simulation and experimental curves of the reference capsule (without barriers).

Step 2 : iterations for finding the "apparent solubitity coefficient" K_s^a of steel which gives the best fit between simulation and experimental curves for the capsule equipped with barriers.

Step 3 : calculation of the permeation reduction coefficient C_{pr} of the barrier **in the test geometry**: the ratio of the permeation rate with barriers ϕ_b to the one without barriers ϕ_{wb} which would be observed if the tritium concentration in Pb17LI were kept constant.

$$C_{pr} = \phi_b/\phi_{wb} = K_s / K_s^a$$

Step 4 : derivation of the C_{pr} value of the barrier for a blanket **design relevant geometry**.

$$C_{pr}^{blanket} = 1 + (C_{pr}^{libretto} - 1) \text{ x } G^{libretto}/G^{blanket}$$

with $G = r_i \text{ Log } (r_i/r_x)$, r_i and r_x being the inner and outer radii of either the LIBRETTO capsule, or the blanket cooling tubes.

5.2 Case with barrier on the downstream side

This case is even simpler, as it can be modelized, as illustrated below, by artificially increasing appropriately the tritium concentration at the capsule wall/sweeping gas interface.

Thus the same procedure as described in section 5.1 can be applied, the only difference being that, in step 2, one will iterate on the "apparent tritium concentration" at the wall/sweep gas interface, instead of on the "apparent solubility" of steel.

6. TENTATIVE APPLICATION OF THE METHOD

The method proposed in section 5.1 was tentatively applied to the case of capsule 8 equipped with an aluminide permeation barrier of 145 µm thickness on the inner surface of its wall. Capsule 6 is here considered as the reference for step 1.

Figure 4 compares the permeation rate history observed experimentally with the results of thre simulations.

The cases where a D_l^y value of 20 times D_l is adopted (the one giving a good fit for the first and final phases of capsule 6 transient) are shown in order to confirm that the transients cannot be explained by an increase in the tritium transport from Pb17Li to the helium plenum. Whatever the reduction made on K_s, the calculated curves do not exhibit the characteristic flat shape observed experimentally.

The case where K_s is reduced by a factor of 75 while D_l^y is kept equal to D_l gives on the contrary a very satisfactory fit with the experiment over the first 180 hours of the transient, until the capsule undergoes a first very small (5°C) temperature transient (two other ones took place at T=218 and 242 hours). Afterwards, simulation and experiment diverge. Globally, such small temperature variations appear to have a significant impact on the permeation rate, indicating the need both to accurately measure and take into account the temperature history.

Although any conclusion would be premature at this stage of the analyses, it seems that the permeation reduction coefficient exhibited by the barrier equipping capsule 8 should turn aroud 80.

7. CONCLUSION

A method relying on the use of a 2D finite element transient simulation model exploiting the analogy between tritium migration and heat conduction phenomena has been developped, which should permit to interpret for the first time the LIBRETTO-type irradiation experiments, among others, in terms of **tritium permeation reduction coefficient** of the tested barriers.

First tests have demonstrated the potentialities of the method and of its associated model. Furthermore, preliminary evaluations seem to indicate that, under conditions similar to the ones of this experiment, the tested permeation barrier could reduce tritium permeation by a factor of about 80.

In the next phase of the study, the analysis of other test runs, especially those with temperature transients and sweep gas transients should permit to further discriminate between the various possible interface laws and boudary conditions. But beyond further analyses model improvements, experimental data on the out-of-pile permeability of steels in presence of Pb17Li are clearly needed to progress in, and validate, the interpretation of such in-pile experiment as LIBRETTO. Such out-of-pile tests are scheduled in 1992 at JRC Ispra.

Another orientation of the future work will be the development of a similar simulation model with a view to help interpret the open capsule irradiation experiment LIPSIE (**Estrade** [7]).

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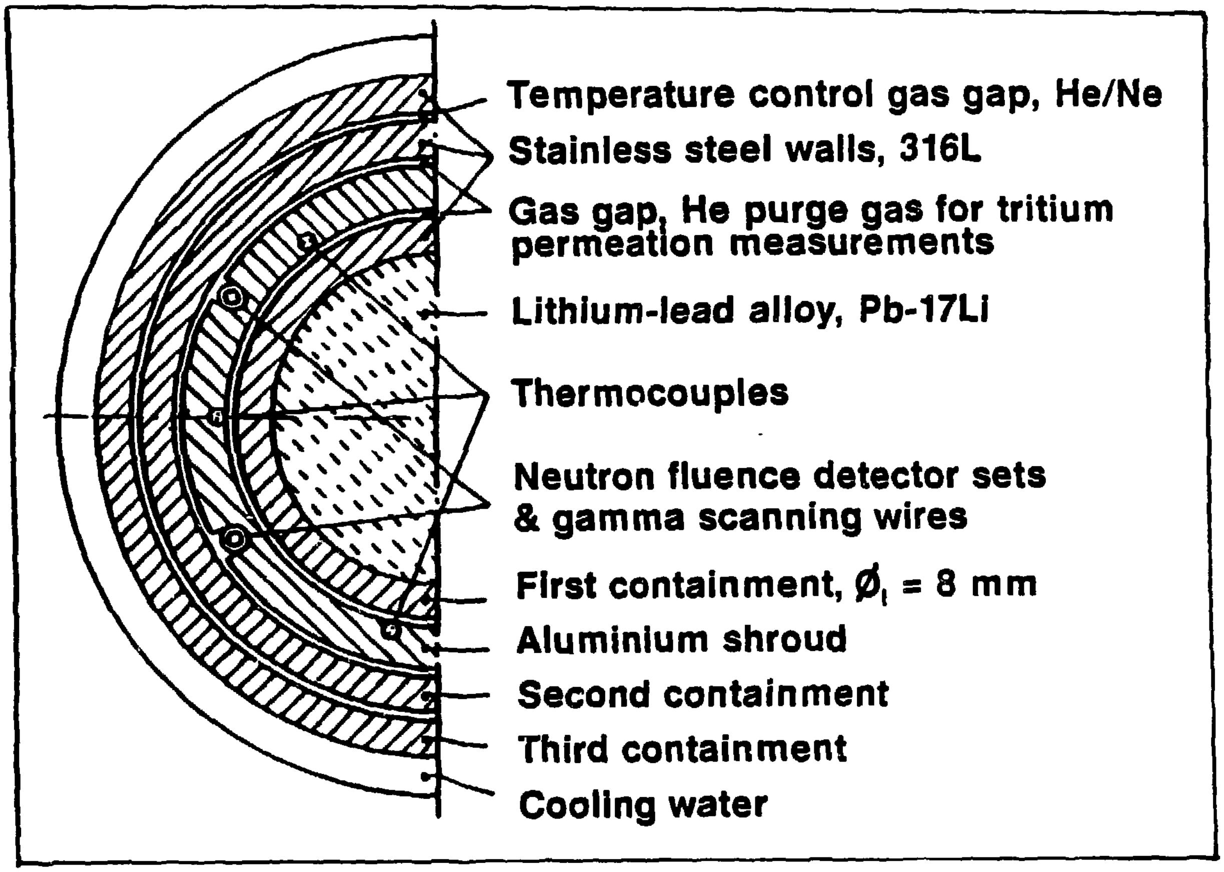

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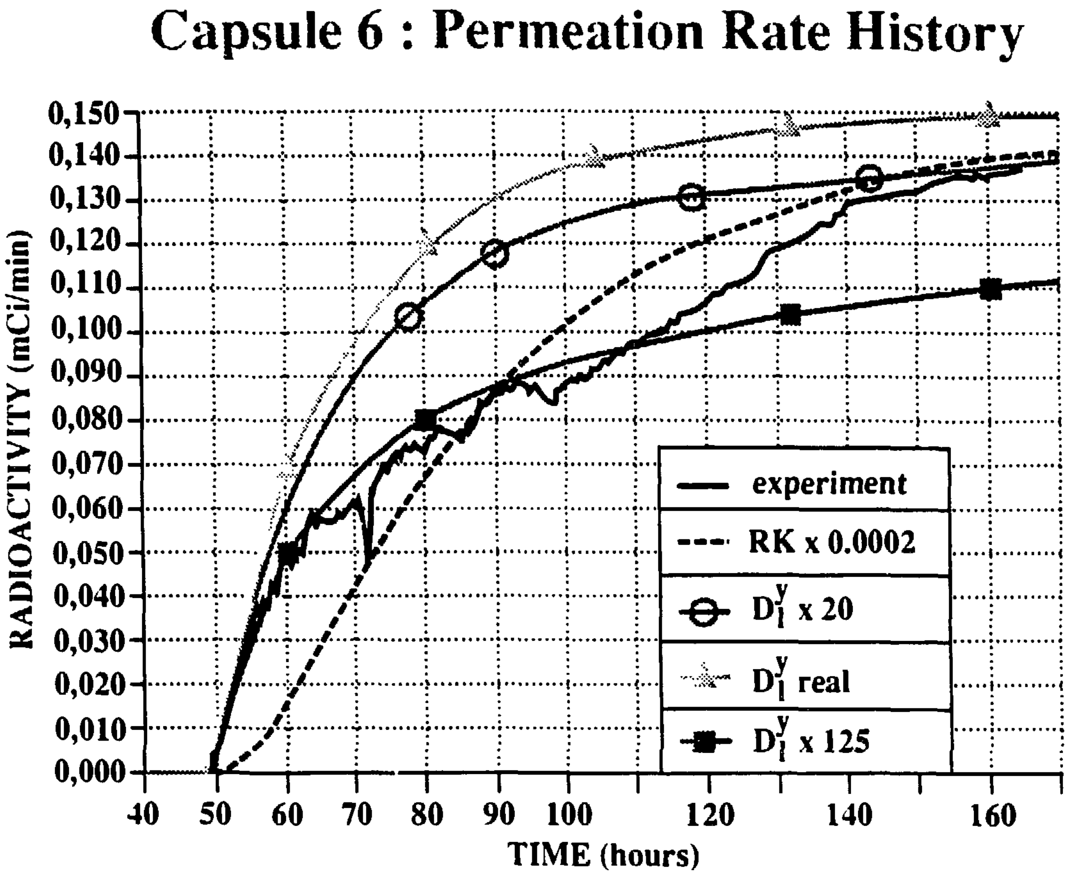

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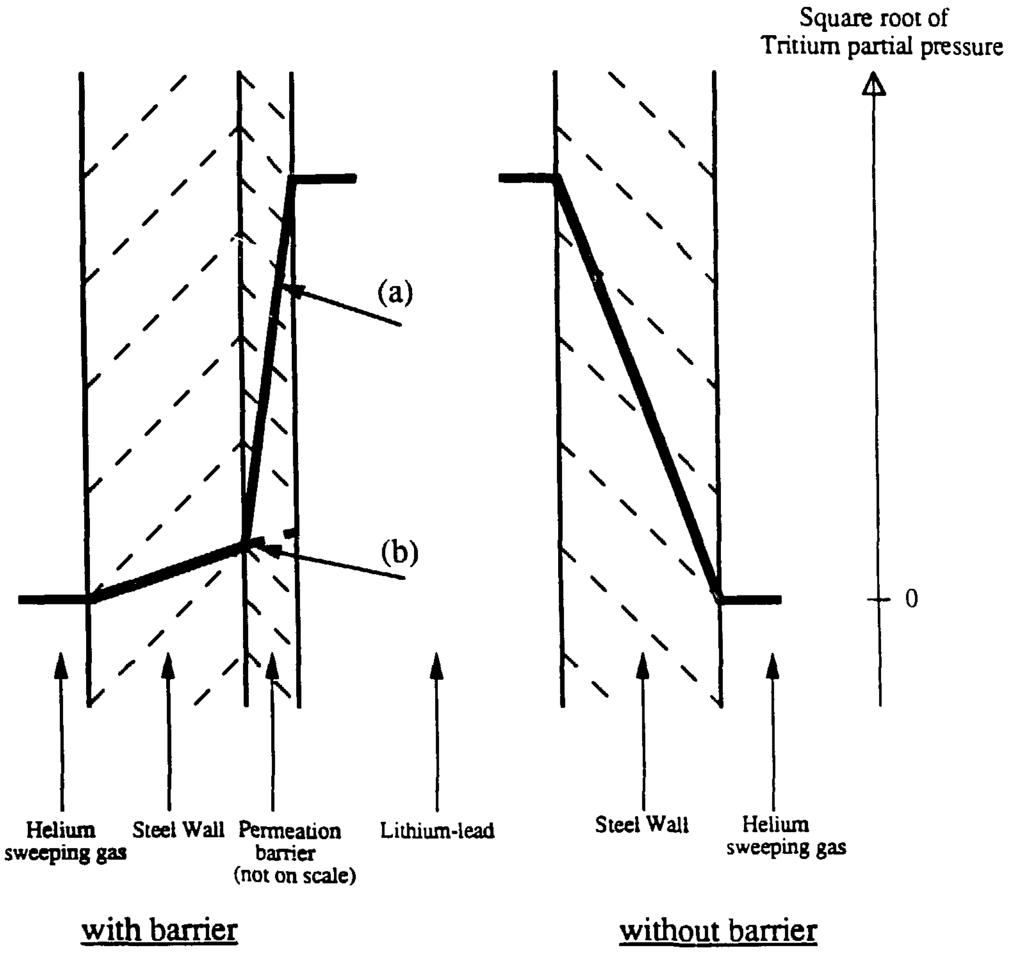

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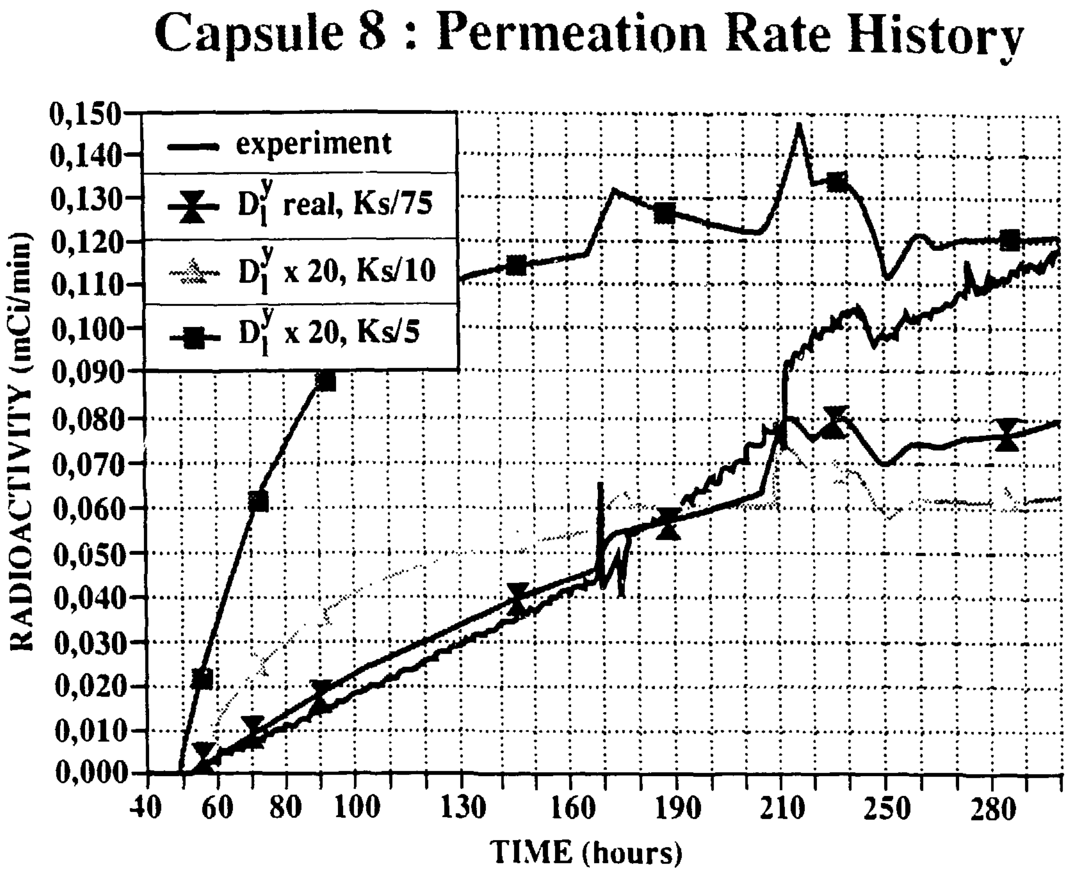

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