MODELLING STABLE-FISSION-GAS DIFFUSION INSIDE THE GRAIN

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ABSTRACT

A new analytical solution for the diffusion of fission-gas atoms inside the UO_2 fuel grain has been obtained. This solution and a related numerical procedure model fission-gas atom migration to the grain boundary under varying conditions of temperature and fission power and including the grain growth process. This new formulation is presented and its improvements and advandages are outlined. The fission-gas release results obtained with a developmental version of ELESIM, which incorporates this new model, as well as some other improvements, are presented.

1. INTRODUCTION

Fission gas release modelling is a major component of any simulation of nuclear fuel behaviour during normal operating conditions, as well as during accidents. This is because of the need to estimate both the source term, and the impact fission gas release has on fuel pin integrity.

Much effort has been devoted to experimental and theoretical studies related to different aspects of fission-gas release, at AECL and elsewhere. A major conclusion of these studies was a general phenomenological model of gaseous fission-gas release from UO_2[1]. There is widespread agreement that there are three main stages before the fission gas atoms reach the free-voidage space.

The first stage involves gas atom migration to the grain boundary, by both diffusion and grain boundary sweeping. Fission gas bubbles precipitate inside the grain and act as traps for diffusing gas atoms. In competition with this process fission gas bubbles also act as gas atom sources because of the re-solution process caused by recoil of fission fragments. As well, the intragranular fission gas bubbles can migrate to the grain boundary under certain conditions of temperature and stress.

The intermediate stage is characterized by the precipitation of grain-face bubbles and the formation of tunnels on the edge of the grains. Diffusion along grain boundaries might also be effective on a short distance scale, but this mechanism is not usually included in fission-gas release models.

In the final stage, the newly created porosity formed by the grain-edge bubbles forms a network of interconnected tunnels with access to open voidage via the outer surface of the pellet or intergranular cracks. This interlinkage of intergranular bubbles occurs at a certain fission-gas atom saturation level of the grain boundary, after which venting of the gas to the free volume occurs. The network of tunnels is maintained as long as enough new gas continues to arrive at the grain boundary. If the network collapses after venting, it takes some time to re-create it.

The purpose of the present paper is to present a new analytical model for gas atom diffusion inside the grain, part of the first stage described above. An outline of the phenomenological model is presented in Section 2. The mathematical model that offers an exact analytical solution and its implementation into a numerical procedure for varying temperature and fission power histories is described in Section 3. Some results obtained by using the new model in the ELESIM code are presented in Section 4, followed by conclusions and plans for future work in Section 5.

2. FISSION-GAS-ATOM MIGRATION TO THE GRAIN BOUNDARY

Fission gas atoms such as Xe, or Kr are not soluble in the UO_2 matrix. They will start diffusing through the matrix, provided the necessary thermo-chemical potential gradient is large enough. The grain boundaries are assumed to be perfect sinks for the fission gas atoms, since once the gas atoms arrive at the grain boundary, they have a higher mobility and are captured by the intergranular bubbles. Therefore, a concentration gradient is created inside the grain, with the grain boundaries at zero concentration. The migration of fission-gas atoms down this concentration gradient is characterized by an intrinsic diffusion coefficient.

The process is complicated by the formation of intragranular bubbles, which act as a distributed sink, as well as a distributed source through the re-solution process. There is still debate on the mobility of intragranular bubbles under various conditions. The treatment of intragranular bubble evolution can be circumvented by using an effective diffusion coefficient, which already incorporates their trapping effect on gas-atom diffusion. This procedure is applicable for normal operating and some abnormal conditions. However, for situations like post-irradiation annealing, when re-solution does not occur, the intragranular bubble concentration must be considered in the model.

Another process that complicates the description of intragranular diffusion is grain growth. By moving through the matrix, the grain boundaries collect the fission gas atoms encountered in their path. The grain boundaries move such that the bigger grains grow at the expense of the smaller grains, which practically disappear after a period. There is actually a distribution of grain sizes, and the grain-growth process is very complex and depends on the initial

microstructure, which is subject to a large variability due to differences in manufacturing parameters.

In principle, the diffusion equation could be solved for different grain-size classes, using a moving boundary diffusion model. This would account for the fact that some grains are growing, and some are shrinking. Still, there are conceptual problems, which make the implementation of this multigroup formalism difficult. For instance, a certain grain-size class can contain both growing and shrinking grains, which have different concentration distributions at the beginning of a simulation time-step. Thus, an exact solution that accounts for past history becomes impossible if individual grain-size categories are to be traced back to the beginning of the simulation.

The procedure adopted here is to use an average grain size. Shrinking grains will eventually disappear, such that their gas inventory will be deposited on the grain boundary in any event. This average grain size is readily available through known correlations based on fitting experimental data. The knowledge of the grain growth process is not advanced enough to warrant a more sophisticated treatment based on moving boundaries.

For the usual applications, the model is based on a number of approximations, shared with most of the models reported in the literature[1,2]. The structural unit of the matrix, for the present application, is the average grain. This is assumed to be spherical, and the concentration inside it is calculated to keep track of the gas migration to the grain boundary, unlike other models which calculate an equivalent sphere radius based on surface/volume ratio. The time is discretized according to the power variation, such that constant temperature can be assumed during any time-increment. The grain growth is modeled as a step-function, the grain radius being constant during a given time-step. The grain-boundary sweeping is calculated incrementally based on the average gas concentration during that time-step. The mathematical model based on these assumptions is outlined below.

3. MATHEMATICAL MODEL FOR FISSION-GAS-ATOM DIFFUSION INSIDE THE GRAIN

The diffusion of gas atoms to the grain boundary can be described by the following equation, treating the grains as spheres of radius a:

$$\frac{\partial C(r,t)}{\partial t} = \frac{D}{r}\frac{\partial^2 [rC(r,t)]}{\partial r^2} + \beta(t) \qquad (1)$$

where C(r,t) is the gas atom concentration, D is the effective diffusion coefficient, which depends on the local temperature, and ß is the source term, calculated as the volumetric fission rate multiplied by the fission yield of fission-gas atoms.

As the diffusion coefficient and the source term vary with time according to the changing power history, the approach taken is to solve equation (1) for a given time-increment, with all

the parameters being treated as constant during the increment. Therefore, the initial and boundary conditions are:

$$\begin{aligned} &C(r,0) = C_0(r) \\ &C(a,t) = 0 \\ &C(0,t) = finite, \frac{\partial C(0,t)}{\partial r} = 0 \end{aligned} \qquad (2)$$

where C_0 is the concentration of gas atoms at the beginning of the time increment, while the grain boundary is considered as a perfect sink as described by equation (2), and t is the time elapsed from the beginning of the time-increment.

To solve equation (1), the dependent variable is changed to W(r,t) , which is then written as the sum of two parts according to the following relations:

$$C(r,t) = \frac{W(r,t)}{r} - \frac{\beta r^2}{6D} \qquad (3)$$

$$W(r,t) = U(r) + V(r,t) \qquad (4)$$

where U(r,t) satisfies the steady-state equation :

$$\begin{aligned} &\frac{d^2U}{dr^2} = 0 \\ &U(0) = 0 \\ &U(a) = C_a = \frac{\beta a^3}{6D} \end{aligned} \qquad (5)$$

while V(r,t) is the solution of :

$$\begin{aligned} &\frac{\partial V}{\partial t} = D\frac{\partial^2 V}{\partial r^2} \\ &V(0,t) = 0 \\ &V(a,t) = 0 \\ &V(r,o) = rC_0(r) - U(r) + \frac{\beta r^3}{6D} \end{aligned} \qquad (6)$$

The solution to equation (5) is immediate, while equation (6) is solved through separation of variables. The final solution is written in the form of a trigonometric series as follows:

$$C^n(y,t) = \frac{\beta_n(1-y^2)}{6D_n} + \frac{2\beta_n}{\pi^3 D_n y}\sum_1^\infty \frac{(-1)^k}{k^3} e^{-\mu_k^2 D_n t} \sin \mu_k y + \frac{2}{y}\sum_1^\infty e^{-\mu_k^2 D_n t} \sin \mu_k y A_k^n \qquad (7)$$

where

$y=r/a$ - normalized radius (8)

$D'=D/a^2$ -effective diffusion coefficient (9)

$\mu_k=k\pi$ (10)

The expansion coefficients A_k^n contain the information related to the fission-gas-concentration distribution at the beginning of the time interval. A recursive relation is obtained for these coefficients, which allows the exact calculation of the varying temperature history to be performed:

$$A_k^n = \frac{\beta_{n-1}(-1)^k}{D'_{n-1}\pi^3 k^3}[e^{-\mu_k^2 D'_{n-1}T_{n-1}} - 1] + A_k^{n-1} e^{-\mu_k^2 D'_{n-1}T_{n-1}} \quad (11)$$

This form of the solution is used to derive the flux of gas atoms to the boundary. After integrating this flux for the respective time step, the percentage release is obtained. This can be split up into two terms that can be related to the old gas and the new gas respectively, such that the absolute fission-gas release for a given time-step can be calculated and added to the previous fission-gas release.

The grain boundary sweeping can be estimated by calculating the swept volume corresponding to the number of grains remaining at the end of the time-step. Alternatively, a balance for the average grain concentration can be written:

$$\frac{dC_{av}}{dt} = -\frac{3}{a}\frac{da}{dt}C_{av} \quad (12)$$

The end result of both approaches is a fractional gas release due to sweeping equal to:

$$f_{sw} = 1 - (\frac{a_0}{a})^3 \quad (13)$$

4. RESULTS

This model was implemented in the fission-gas release module in a developmental version of the ELESIM code and is currently being benchmarked, other improvements to the fission-gas release model addressing the second and the third stages described in Section 1, were presented previously[3].

The experiments in the fission-gas release database[4] were simulated and the results showed improved agreement between the measurements and the simulations. The fission-gas releases calculated with both the current and the revised(developmental) versions of ELESIM are compared with the experimental data in Figures 1 and 2 for the normal burnup range, and in Figures 3 and 4 for the extended burnup data range.

Since this model eliminates the time-step sensitivity, there are now no limitations on the burnup increment size and number. The main improvements are that the high-burnup fission-gas release data are better reproduced, and that the previous wide spread of the calculations is reduced significantly, showing a more consistent response of the code. This reduction in the variance between the calculations and the measurements is attributed to the new diffusion model presented in this paper.

5. CONCLUSIONS

A model for diffusion of stable fission gases inside the UO_2 fuel grain has been defined, which is applicable to normal and abnormal operating conditions. The main improvement of the present treatment is the exact solution to the diffusion equation inside the grain with varying diffusion coefficients and a moving boundary. This model is limited only by the assumption that the diffusion coefficient and grain boundary position remain constant during a time-step.

The analytical solution used previously[2], lacked precision because of the assumed uniform initial concentration for any time-step (relation 2), which made the calculation very sensitive to the number of time-steps used. The analytical solution presented here has the added advantage of being fast and robust, without any of the problems that the numerical methods might experience.

The present formulation of the model is considered precise from the theoretical viewpoint, and sufficiently accurate with regard to the approximations introduced to model the real process. Future work will focus on implementing the model for the radioactive-fission-gas release needed for accident calculations.

6. REFERENCES

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(3) V.I. ARIMESCU and W.R. RICHMOND, "Modelling CANDU-type Fuel Behaviour During Extended-Burnup Irradiations Using A Revised Version of the ELESIM Code", AECL report AECL-10622, 1992.

(4) W.R. RICHMOND, J.M. BUNGE and V.I. ARIMESCU, "Extended-burnup CANDU Fuel: Description of Database and Comparisons with Code Predictions", AECL report AECL-10655, 1992.

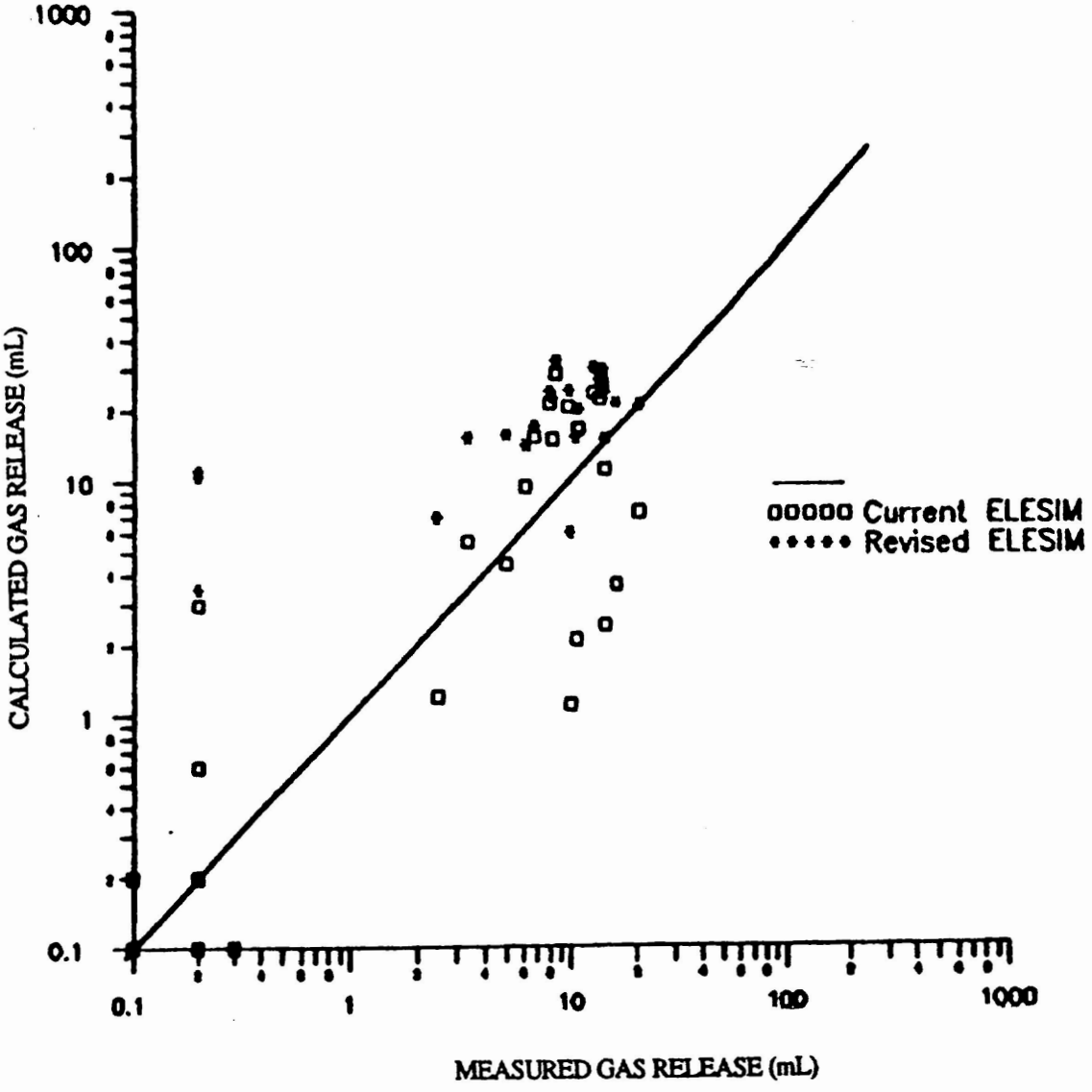

FIGURE 1: COMPARISON BETWEEN MEASURED AND CALCULATED FISSION-GAS RELEASES USING THE CURRENT AND THE REVISED VERSIONS OF ELESIM FOR THE POWER-REACTOR NORMAL BURNUP DATABASE.

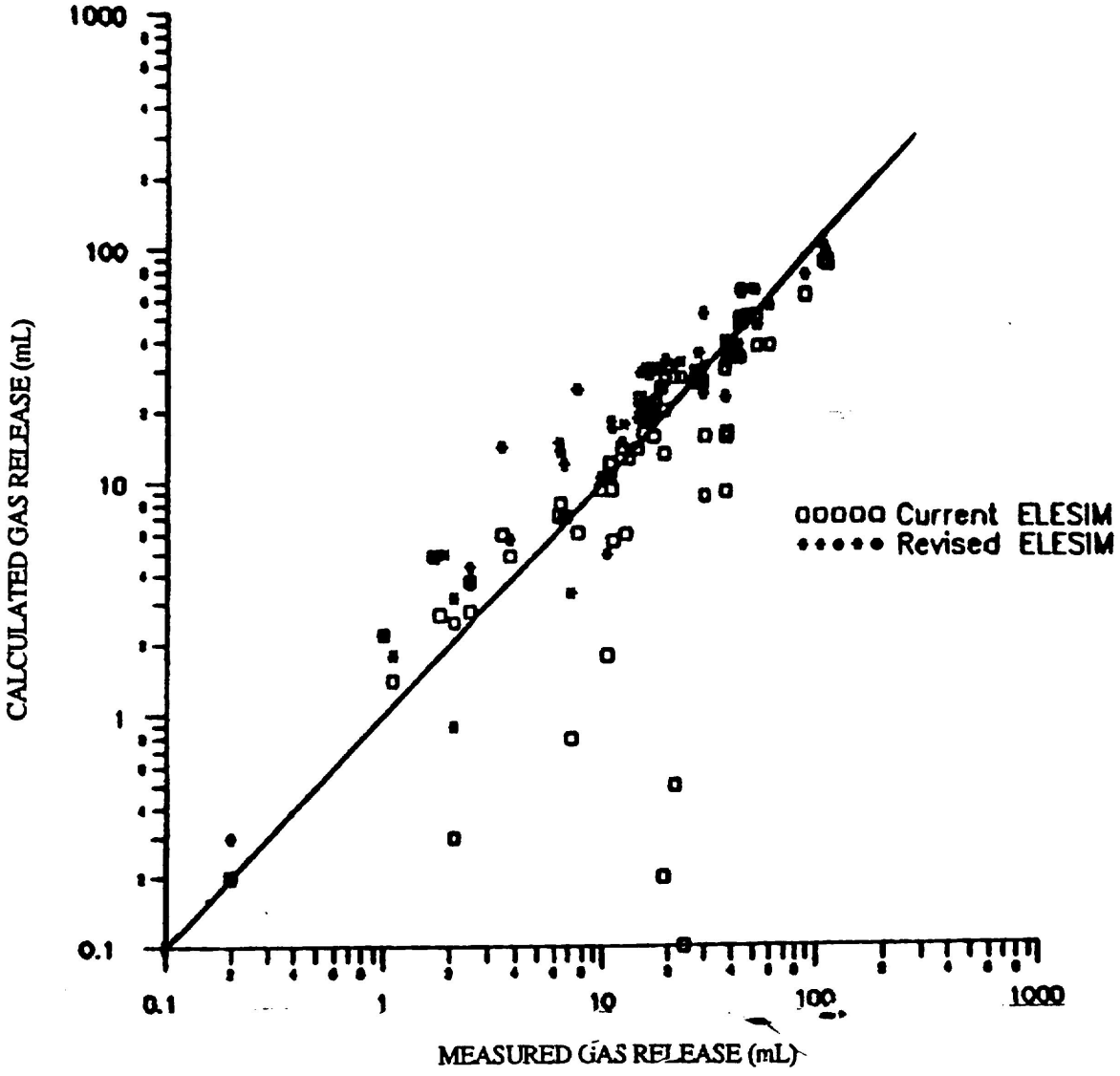

FIGURE 2: COMPARISON BETWEEN MEASURED AND CALCULATED FISSION-GAS RELEASES USING THE CURRENTAND THE REVISED VERSIONS OF ELESIM FOR THE RESEARCH-REACTOR NORMAL BURNUP DATABASE.

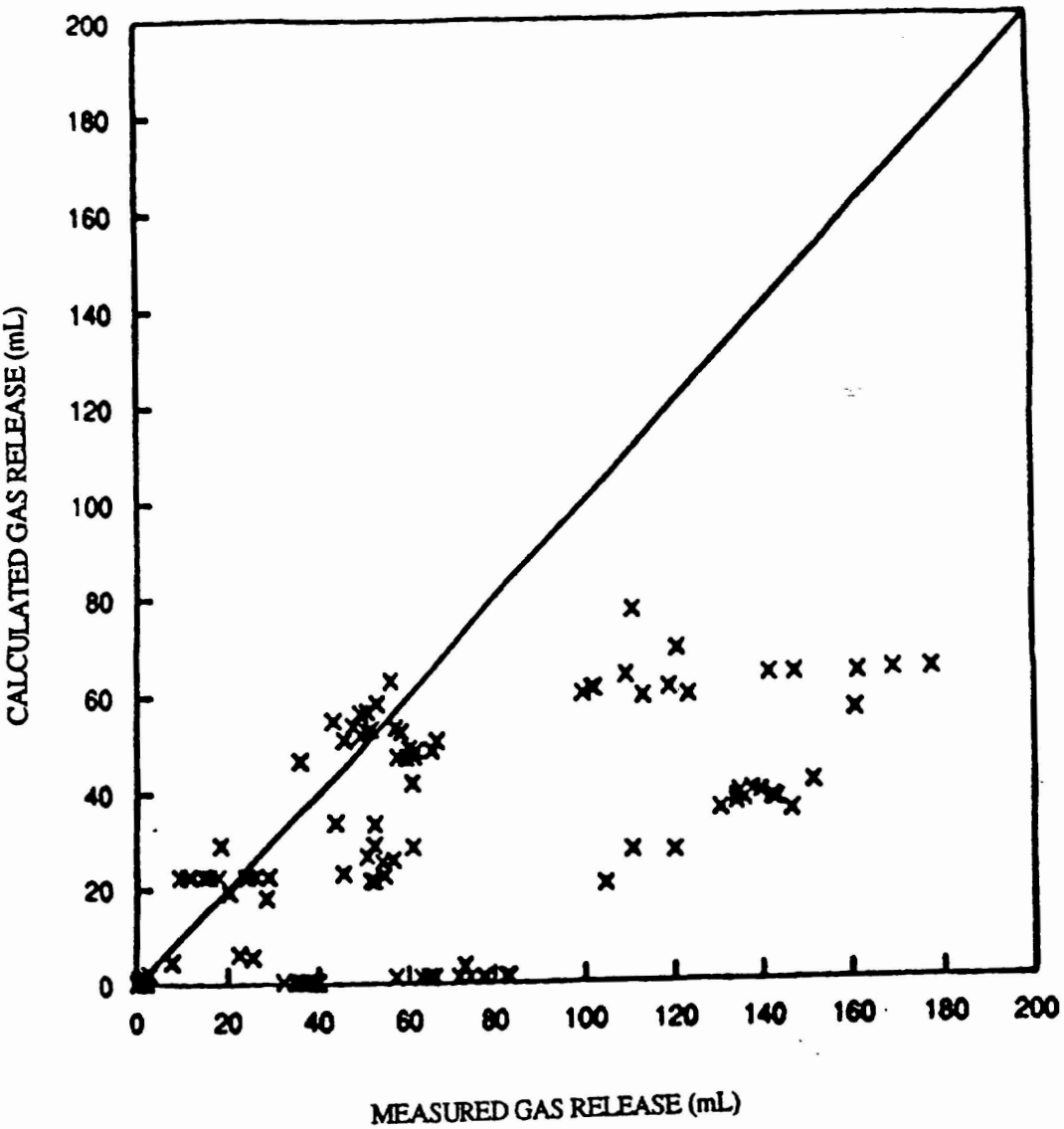

FIGURE 3: COMPARISON OF THE CURRENT VERSION OF ELESIM CALCULATION OF FISSION-GAS RELEASE AGAINST THE EXTENDED BURNUP DATA.

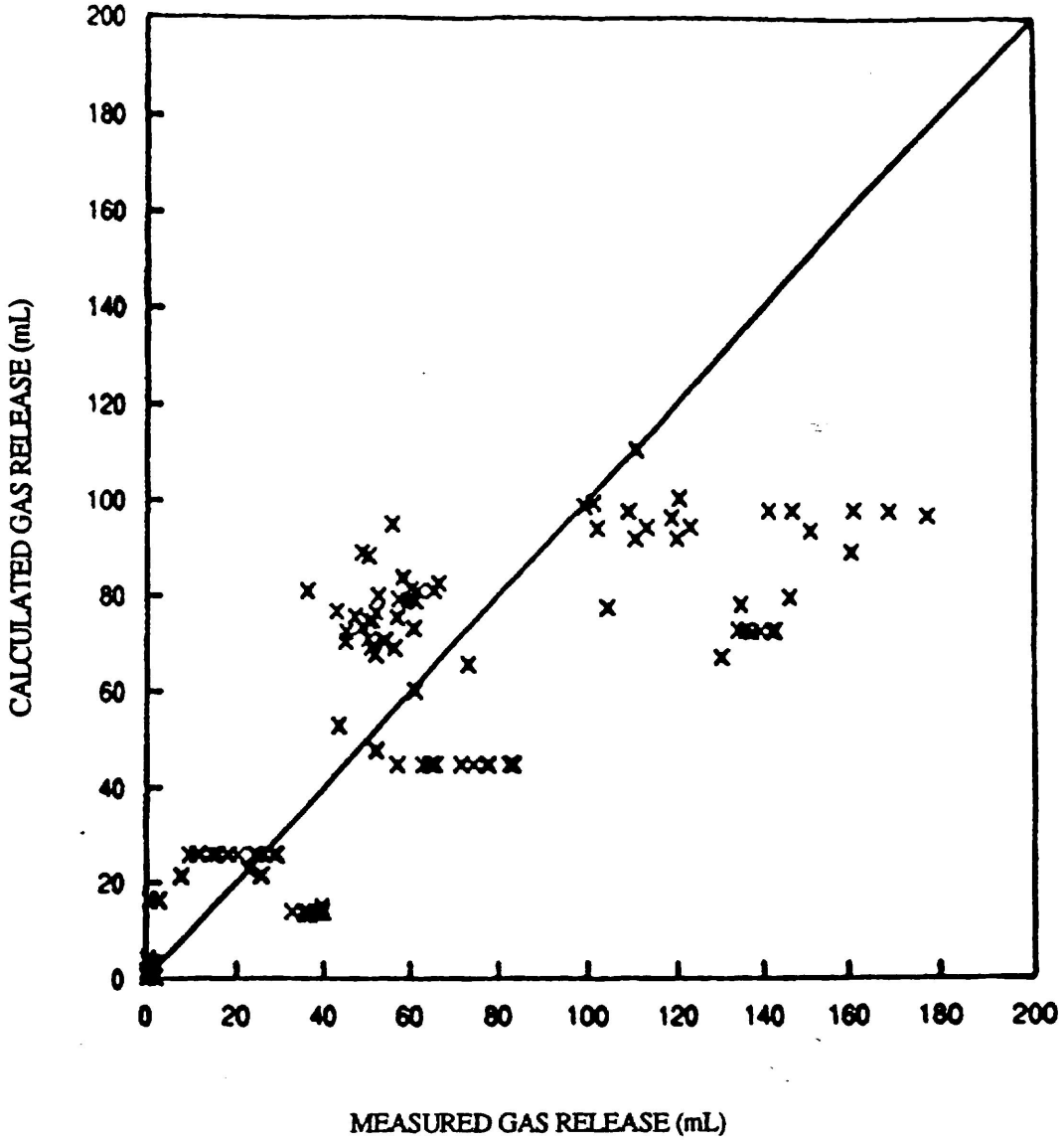

FIGURE 4: COMPARISON OF THE REVISED VERSION OF ELESIM CALCULATION OF FISSION-GAS RELEASE AGAINST THE EXTENDED BURNUP DATA.