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An Improved Model for the Release of Fission Gas in CANDU Fuel

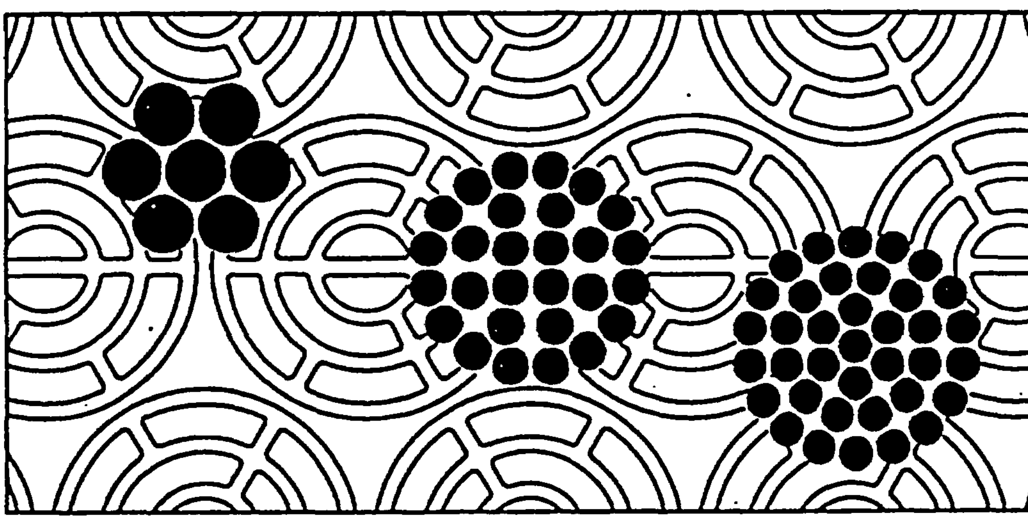
by M. Tayal and A. Ranger

presented at:
**The Fifteenth Annual
Nuclear Simulation Symposium,
Mississauga, Ontario
May 1-2, 1989**

AECL-9798

May 1989

FUEL BRANCH



SAFETY ENGINEERING DEPARTMENT

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SUMMARY

Previous studies have indicated the desirability of increasing the burnup of CANDU fuel, from 7-9 GWd/t to about 21 GWd/t. At extended burnups, one issue in fuel integrity is fission gas release. Earlier studies showed that at extended burnups, fuel performance codes ELESIM [1] and ELESTRES [2] underpredict the release of fission gas by about a factor of two. An improved model is proposed in this paper.

Via parametric studies that compared measurements to predictions, ad-hoc modifications have been determined for the following sub-models affecting gas release: thermal conductivity of UO_2; diffusivity; and density of intergranular bubbles. The levels of the modifications are consistent with experimental data.

The new model improves the predictions of ELESTRES for fission gas release when compared against measurements from 98 irradiations in research as well as in commercial reactors. On average, the predictions now differ from the measurements by 1.3%. For reference, the experimental scatter in a typical irradiation is ±3-4%.

UN MODELE AMELIORE DU DEGAGEMENT DES GAZ DE FISSION DANS LE COMBUSTIBLE CANDU

Par

M. Tayal et A. Ranger

Présenté au 15e Symposium Annuel sur la Simulation Nucléaire

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RESUME

Les études ont démontré qu'il est désirable d'augmenter le taux de combustion du combustible CANDU de la valeur présente (7–9 GW.j/t) à environ 21 GW.j/t. Aux valeurs élevées de combustion massique, une question qui porte sur l'intégrité du combustible est le dégagement des gaz de fission. Des études antérieures ont montré qu'aux valeurs élevées de combustion massique, les logiciels ELESIM(1) et ELESTRES(2) de performance du combustible sous–estiment le dégagement des gaz de fission par un facteur de deux. Cette communication propose un modèle amélioré.

Des études paramétriques comparant les mesures aux prédictions ont servi à faire des modifications ad hoc aux sous–modèles suivants touchant au dégagement des gaz: la conductivité thermique de l'UO_2; la diffusivité; et la densité des bulles intergranulaires. La grandeur des modifications est compatible avec les données expérimentales.

Le nouveau modèle améliore l'accord entre les prédictions d'ELESTRES sur le dégagement des gaz de fission et les mesures provenant de 98 irradiations en réacteurs de recherche et réacteurs commerciaux. La différence entre les prédictions et les mesures est à présent de 1,3% en moyenne. Par comparaison, la dispersion des résultats d'une irradiation typique est de ±3–4%.

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1. INTRODUCTION

Atomic Energy of Canada Limited (AECL) has an ongoing program to maintain and improve the competitive position of CANDU by improving resource utilization and by reducing unit energy costs. The use of slightly enriched uranium (SEU) in CANDU has been identified as offering substantial returns. Use of SEU improves resource utilization and reduces unit energy costs, via reduced fuel cycle costs. There is an increase in discharge burnup for SEU over natural uranium (NU) which exceeds the increase in uranium feed requirements; there is also a corresponding decrease in back-end, spent-fuel volume.

The designers of CANDU-6 Extended Burnup Core (EBC) have concluded that a core-average exit burnup of 21 GWd/tU is desired, taking into account burnup penalties due to the end flux suppressor and burnable poison. In comparison, the current CANDU reactors have core-average discharge burnups of 7-9 GWd/tU. The burnup target, which is about three times that for the current CANDU-6 natural uranium fuel, plus changes in some operating conditions, place increased demands on fuel performance.

One of the issues in fuel integrity is the additional gas release at extended burnups. It was noted that the fission gas package, NOTPAT, of the fuel performance codes ELESIM [1] and ELESTRES [2] underpredicts gas release by about a factor of 2 at extended burnups.

This suggests a need to improve the gas release predictions, because gas release can have important consequences on fuel integrity during normal operation as well as during hypothetical accidents involving high temperatures (such as Loss of Coolant Accidents).

It is expected that the internal gas pressure in the CANDU-6 EBC fuel will stay below the coolant pressure during normal operation. Thus the sheath and the pellet will stay in radial contact, i.e. 'lift-off' is not expected. For this reason, the improvements to the fission gas model were limited to conditions before lift-off.

This paper first describes the mechanisms contributing to fission gas release. Then, the method used to obtain the new model is described, followed by a discussion of the new model. The predictions of the new model are compared with measurements from about 100 irradiations in research and in power reactors.

2. MECHANISMS

2.1 Effects of Gas Release

Fission gas release is important for the following reasons:

a) During high-temperature transients such as a Loss of Coolant Accidents, the gas pressure in the fuel element is the major driving force for sheath expansion and possible failure via ballooning/overstrain.

b) During normal operation, the fission gas pressure is designed to stay below coolant pressure, keeping the sheath in contact with the pellet. However, if the gas pressure were to increase much above the coolant pressure, the gas may force the sheath away from the pellet; this is called lift-off. This can reduce the heat transfer coefficient between the pellet and the sheath, increasing the pellet temperatures, and promoting further increase in gas release and in gas pressure. The feedback can potentially lead to an unstable situation, ultimately resulting in sheath failure via excessive stress/strain. To avoid this, the gas pressure is kept within acceptable limits.

c) Positive differential pressure (gas minus coolant) will also impose tensile stresses on the sheath, which can promote the migration of hydrogen/deuterium to regions of high stresses, making the sheath more vulnerable to failure via environmentally-assisted cracking in a subsequent power-ramp. The tensile stresses due to gas overpressure will also add to the tensile stresses due to pellet expansion (hourglassing, fission product swelling), posing additional risk to sheath integrity.

d) Some fission products, like iodine and cesium, promote stress-corrosion cracking of the sheath. Their concentration at the sheath surface should be kept low, for example via CANLUB and via low release of fission gas.

e) Knowledge of gas release can be used during design to avoid the problems identified above.

2.2 Steps During Gas Release

Figure 1 illustrates the terms used to describe the gas release model. The space between the pellet and the sheath is often called the 'open void'.

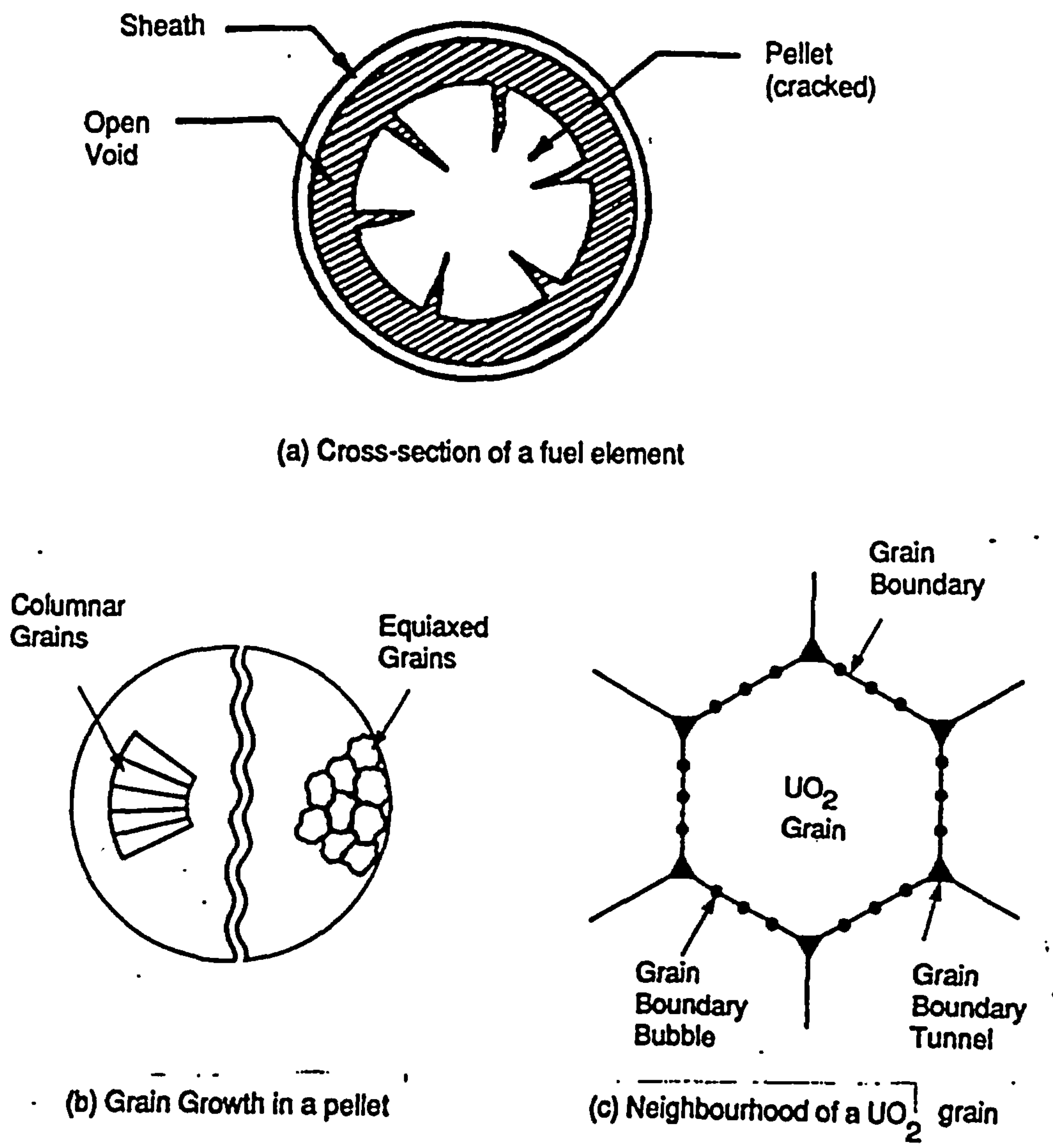


FIGURE 1: ILLUSTRATION OF SOME TERMS USED IN DESCRIBING THE MODEL

Irradiation generates fission products within the interior of UO_2 grains. Some of the fission products are gaseous. The gas diffuses through the UO_2 grains to the grain-boundaries, due to differences in local concentrations of the gas [1].

The diffused gas amasses in bubbles at the grain boundaries. The bubbles grow as more gas reaches the grain boundary, either by diffusion or by grain-boundary sweeping due to grain growth. When the bubbles are big enough to touch each other (i.e. to interlink), any excess gas in the bubbles is assumed to be available for release to the grain-boundary tunnels. From the tunnels, it is released to the open void at the next change in reactor power. If the UO_2 temperature exceeds the UO_2 melting point, then the gas contained within the grains and within the grain-boundaries of the affected areas, is released directly to the open void.

Table 1 shows the parameters which have the most influence on the above processes. They include: pellet temperatures; heat transfer coefficient between the pellet and the sheath; local heat generation rate as a function of distance along the pellet radius and of burnup; thermal conductivity of UO_2; rate of grain growth; efficiency of grain boundary sweep; density of intergranular bubbles; fraction of grain surface covered by intergranular bubbles; and frequency of power changes. Of these, we focused on the roles of the following three parameters, because experimental evidence suggests [3-17] their dependence on burnup:

- thermal conductivity of UO_2
- diffusivity
- density of intergranular bubbles.

The following paragraphs describe the three in more detail.

TABLE 1: <u>PARAMETERS RELEVANT TO FISSION GAS RELEASE</u>

STEP	PARAMETER	TUNED?
1. DIFFUSION	TEMPERATURE	
	- HEAT TRANSFER COEFFICIENT BETWEEN THE PELLET AND THE SHEATH	
	- HEAT GENERATION RATE	
	- THERMAL CONDUCTIVITY	√
	DIFFUSIVITY	√
2. GRAIN BOUNDARY SWEEP	- RATE OF GRAIN GROWTH - EFFICIENCY OF SWEEP	
3. STORAGE IN BUBBLES AT GRAIN BOUNDARY	- BUBBLE SIZE	√
	- FRACTION OF GRAIN SURFACE COVERED BY BUBBLES	
4. RELEASE TO OPEN VOID	- FREQUENCY OF POWER CHANGE	

2.3 Thermal Conductivity of UO_2

The thermal conductivity of UO_2 has been thoroughly investigated. Most of the correlations suggested in the open literature account for the dependence on temperature, on density, on fission product swelling, and on stoichiometry, whereas effects of impurities and of burnup are usually neglected. ELESTRES uses the MATPRO-11 correlation [3], which is also based on measurements of thermal conductivity in unirradiated UO_2. The effect of burnup is incorporated via its effect on density and on fission product swelling. For CANDU fuel, the correlation gives thermal conductivities of 2-5 W/(m.K). The uncertainty in the correlation has a standard deviation of 0.35 W/(m.K).

It has been postulated [4] that irradiation can change the thermal conductivity of UO_2 via the following influences:

- New isotopes and elements are produced during the irradiation;
- Fission gas bubbles are produced within the grains of UO_2 and on the grain boundaries;
- Macro and micro-cracks are created in the pellet, and
- The stoichiometry of the pellet changes.

Namekawa et.al report [5] that irradiation to 700-800 MW.h/kg decreases the conductivity of (U, Pu)O_2 pellets by about 0.5 W/(m.K). Although they state that the decrease is within experimental error, the shift is remarkably consistent with temperature, and similar to that reported by other experimenters.

Kleykamp reports [6] that at 10% burnup the solid solutions formed during the irradiation decrease the thermal conductivity by 19-28% at 1000-2000 K, i.e. by about 0.4-1 W/(m.K). He offers the following explanation:

"The thermal conductivity of oxide fuels changes during irradiation. It deteriorates due to the formation of a solid solution of the fuel with the oxides of zirconium, strontium, the rare earths and other fission products in lower concentrations and is not counterbalanced by the metallic and oxide fission product inclusions precipitated within the fuel and in the grain boundaries".

Balancing the above, opinion has also been expressed that enhanced gas release at high burnup is not due to significant changes in thermal conductivity but due to, for example:

- saturation of intragranular bubbles, in that the number of gas atoms in the bubbles becomes equal to the bubble capacity [7];
- faster grain growth [8].

Turnbull's arguments about bubble saturation [7] are based on analysis of Vitanza's measurements [9]. Baron-Maffeis' conclusions about grain growth [8] are based on a reanalysis of the FRAGEMA data-base. In addition, Lassman compares [4] the predictions of the TRANSURANUS code to one measurement of pellet centreline temperature using thermocouples, and concludes that the "effect of burnup on the thermal conductivity is not significant" for burnups < 720 MW.h/kgU.

At this time, it is hard to determine from first principles precisely how the thermal conductivity changes with burnup due to the combined influences of all the pertinent effects. Direct measurements of the thermal conductivity of irradiated material are sparse, as are thermocouple measurements at high burnups. Even when the latter are available, they are difficult to interpret because of the uncertain thermocouple decalibration during the irradiation and because of the uncertainty originating from the uncertain heat transfer coefficient between the pellet and the sheath at high burnups.

Widespread concerns persist [10] that the changes in thermal conductivity may enhance gas release at high burnups. For this reason detailed programs are underway [10] to define this effect more precisely.

In the meantime fuel modelling codes often employ normalization factors to account for the effects of irradiation on pellet temperature. For example, the FRAPCON code [11] contains an empirical factor, called either R or CFAC in FRAPCON, which reduces the conductivity of UO_2. For conditions in CANDU fuel, it is estimated that the correlation in FRAPCON would predict that the 'effective' thermal conductivity of UO_2 is a factor of 2-3 lower than that given by the MATPRO correlation. Similarly, the FEMAXI code [12] assumes that the 'effective' surface roughness of the LWR pellets is 4 µm, four times higher than the real roughness.

The reference version of ELESTRES, called the M11C version, does not contain similar normalizations related to temperature calculations.

2.4 Diffusivity

The literature suggests [13-16] that irradiation can sometimes decrease and sometimes increase the diffusivity of Xe in UO_2.

Based on measurements, Matzke reports [13] that depending on the local temperature in the range 1400-1550°C, the diffusivity of gas in UO_2 after 2.0 to 2.6 x 10^{17} fissions/cm³ is 3 to 20 times lower than that measured after 5 to 8 x 10^{14} fissions/cm³. MacEwan and Stevens also measured [14] decreases in diffusivity with increasing irradiation exposure. These results indicate that the gas atoms are trapped by irradiation - induced defects, resulting in retarded diffusion.

On the other hand, some experimenters have suggested that impurities, new isotopes, and changed stoichiometry can increase the diffusivity by up to a factor of 50 [15].

With irradiation (fission), some of the original U in UO_2 gets converted to lighter atoms, giving an 'excess' of oxygen, i.e. UO_{2+x}. But some of the lighter fission fragments use oxygen. Also, early experiments at U.S. G.E. showed that oxygen has affinity to Zr, leaving the stoichiometric composition of the pellet virtually unchanged. Recently, however, Walker and Mogensen show [16] build-up of oxygen at the centre of the pellet in one fuel rod (two were examined), which at first glance does not appear consistent with the early experiments at GE. The stoichiometry is further influenced by the stability of the lighter oxides at the operating temperatures. The complicated dynamics of the stoichiometry in a fuel element can only be known by a detailed chemical model; none are available at present. At this stage, it is therefore difficult to predict [16] the stoichiometry within an operating fuel element.

2.5 Density of Grain Boundary Bubbles

ELESIM and ELESTRES assume that the density of intergranular bubbles is 6×10^8 bubbles per square cm of the grain boundary area [1]. This means that the bubbles interlink when they attain, on average, a radius of 0.23 μm. This number is considered in ELESTRES to be a constant, and therefore independent of parameters like: temperature; surface tension; burnup; quantity of fission gas on the grain boundary; etc. The basis of the assumption comes from post-irradiation examinations of 6-10 fuel bundles, all irradiated in the Pickering reactor to burnups of 190-220 MW.h/kg, at linear heat generation rates of 44-50 kW/m.

Considering that the above data are sparse, it is conceivable [18, 19] that at other operating conditions the bubble density is a function of local parameters like the ones mentioned above. As an illustrative example, bubble radii of about 5-10 μm have been reported by Turnbull [18, 19] and by Kleykamp [6]. Hastings et al. have also observed bubble radii ranging from about 0.05 to 5 μm from an electron fractograph in irradiated fuel from Pickering [20]. Moreover, reference [2] shows that in CANDU fuel the bubble-density at high burnups (∿ 700 MW.h/kgU) is a factor of 10 lower than at moderate burnups (∿200 MW.h/kgU). This means that the bubble radius for interlinkage can be a factor of 10 higher at high burnups than at moderate burnups. This has been a known limitation in the gas release model of ELESIM/ELESTRES: "...Deficiencies in the gas release model stem from its empirical nature. The role of gas bubbles as an intermediate stage in release is important as it is probably the reason for the burnup-dependency of release rates [19]. No such dependence is modelled into ELESIM......"

Recently, KAERI showed [21] that a more sophisticated model using variable bubble density/size reduces significantly the scatter in ELESIM predictions compared to measurements. This package, however, is not yet a part of the AECL-version of ELESIM/ELESTRES.

In summary, we changed the following three parameters to improve the gas release predictions of ELESTRES:

- thermal conductivity of UO_2;
- diffusivity of fission gas in UO_2; and
- density of grain-boundary bubbles.

The method and the results are discussed in the following sections.

3.0 METHOD

CRNL provided us a data-bank of 233 irradiations, containing a maximum burnup of about 700 MW.h/kgU, for which fission gas measurements are available. Of these, 123 irradiations are presently considered qualified for purposes of code validation, because:

- The ELESIM/ELESTRES input data for these irradiations have been reviewed and there is reasonable confidence in their accuracy, and

- These irradiations did not contain significantly atypical conditions that bear on gas release, such as grooved pellets, mixed oxide pellets, etc.

We reviewed the 123 qualified irradiations, and concluded that 25 of them likely contained sheath lift-off. The remaining 98 irradiations were considered valid and used in this study. They include irradiations in research reactors as well as in power reactors.

The irradiations used in this study had a maximum linear heat generation rate of 110 kW/m, maximum burnup of 362 MW.h/kgU, and maximum gas release of 44%. Percent gas release refers to the gas released to the open void as a percent of the gas produced during the irradiation.

The valid irradiations fall in the following ranges of burnups:

0 - 100 MWh/kg: 30 irradiations

100 - 200 MWh/kg: 33 irradiations

200 - 300 MWh/kg: 30 irradiations

300 - 400 MWh/kg: 5 irradiations .

Thus, the valid data are distributed quite evenly in the range 0-300 MWh/kgU, and are sparse above this burnup.

The valid irradiations were simulated using the ELESTRES code, and the predictions were compared to the measurements using two regression models: y = mx; and y = mx + c. Table 2 gives the results. 'Slope' refers to 'm', and 'intercept' refers to 'c'.

TABLE 2: SUMMARY OF PREDICTIONS OF ELESTRES.M11C FOR FISSION GAS RELEASE

Regression Model:		y = mx	y = mx + c	
Burnup	Number of Data Points	Slope 'm'	Slope 'm'	Intercept 'c'
(MWh/kgU)				(%)
0-100	30	0.8551	0.9391	-1.92
100-200	33	0.7032	0.6821	0.48
200-300	30	0.2811	0.2236	1.08
300-400	5	0.5008	0.3958	2.19
All Data	98	0.6856	0.6950	-0.21

Notes: y = predictions
x = measurements
m = slope
c = intercept

The following trends were noticed:

1. The code generally undepredicted gas release, as noted earlier. The undeprediction generally increased with burnup, see Table 2, although the trend was not monotonic. Therefore, the data used in this study do not suggest an abrupt decrease in the thermal conductivity at a critical high burnup. Instead, a gradual decrease is suggested, starting at low burnups. This is qualitatively consistent with the assumptions used in the FRAPCON and FEMAXI codes. It is also consistent with the results of Lassman and of Turnbull. Other interpretations of the data are also possible, such as faster grain growth.

2. The slope first decreases monotonically till 300 MWh/kgU, then there is an increase in slope for 300-400 MWh/kgU. Even though the data are sparse in the latter range, a smoother trend can be established by, for example, using variable density/radius of intergranular bubbles, per KAERI [21].

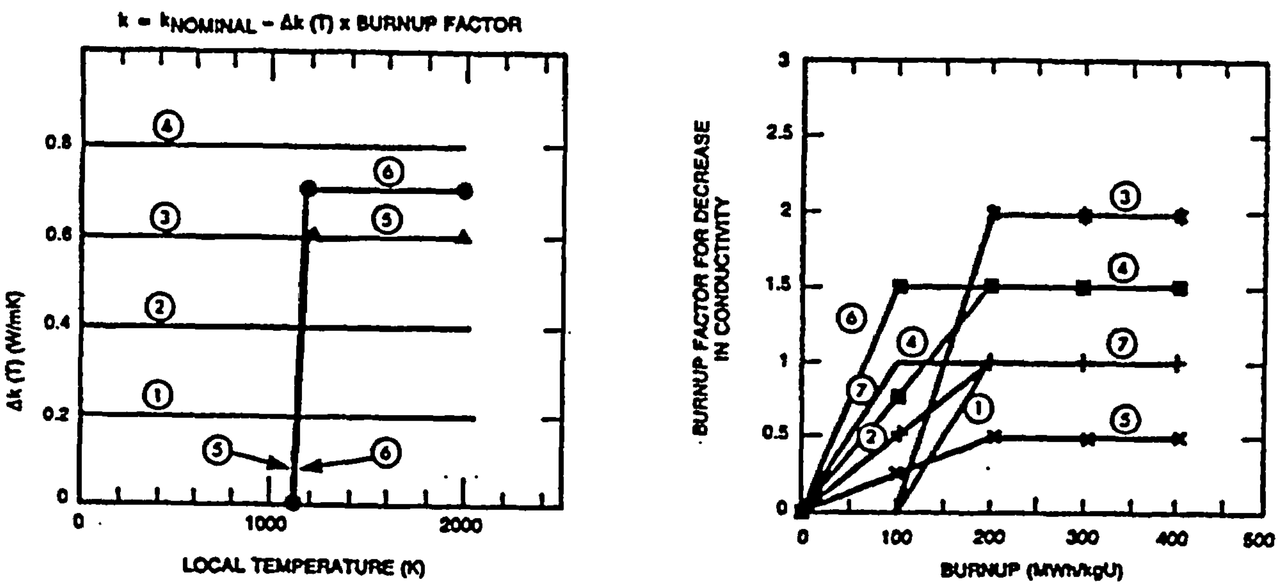
We then tried parametric studies of the following parameters:

a) Decrease in thermal conductivity as a function of temperature: six attempts; see Figure 2a. When the conductivity was decreased by a constant amount (independent of temperature; curves 1-4 of Figure 2a), it was difficult to match the predictions to the measurements over the complete range of gas releases. Therefore, the change in thermal conductivity was then treated as a function of temperature; see curves 5-6 of Figure 2a.

b) Decrease in thermal conductivity as a function of burnup: seven attempts; see Figure 2b.

c) Diffusivity as a function of burnup and of local temperature: three attempts; see Figure 2c.

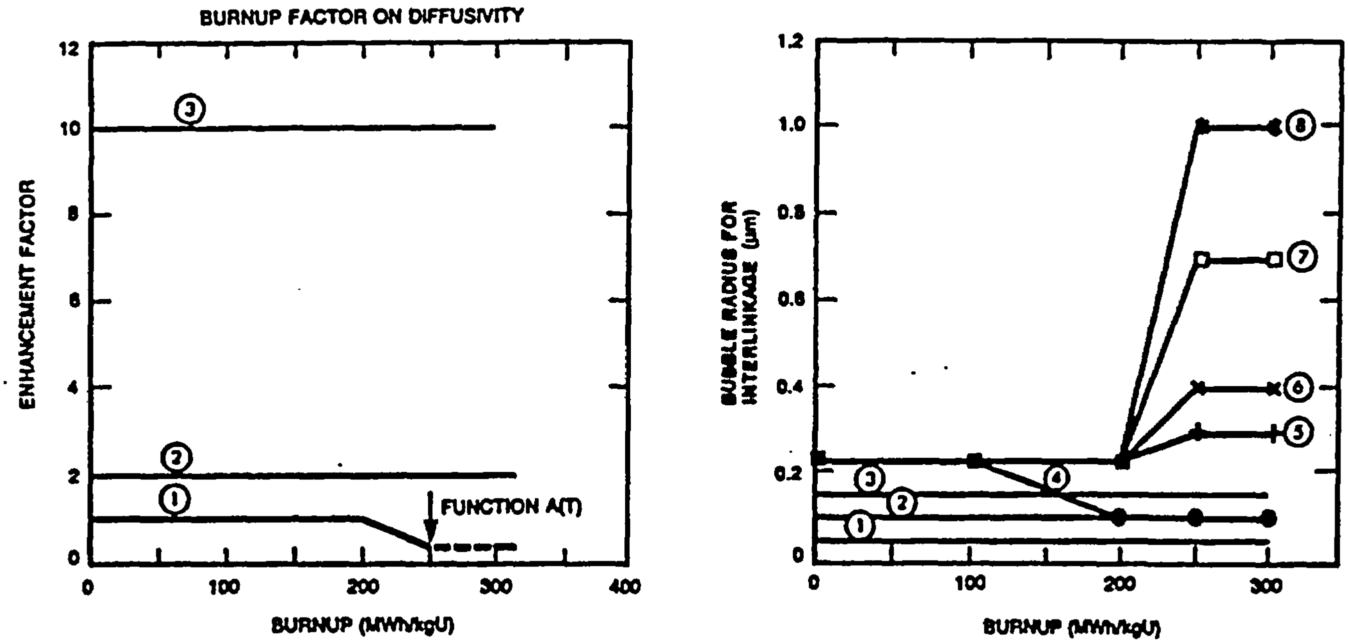
d) Density of grain-boundary bubbles as a function of burnup: eight attempts; see Figure 2d.



(a) THERMAL CONDUCTIVITY AS A FUNCTION OF TEMPERATURE

(b) THERMAL CONDUCTIVITY AS A FUNCTION OF BURNUP

FIGURE 2 ATTEMPTED ALTERATIONS OF PARAMETERS



(c) DIFFUSIVITY AS A FUNCTION OF BURNUP

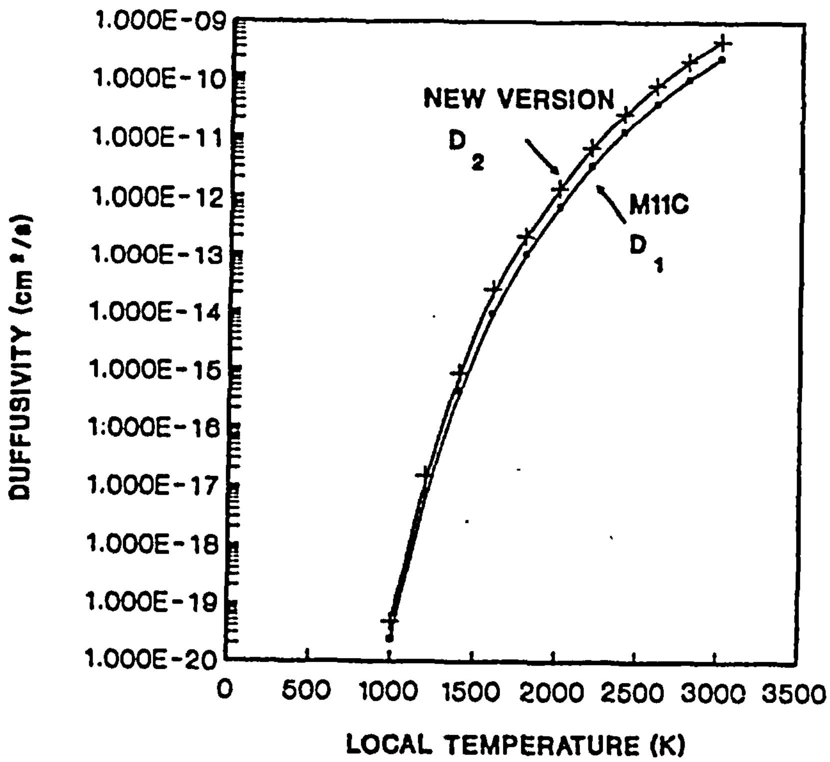
(d) BUBBLE RADIUS FOR INTERLINKAGE AS A FUNCTION OF BURNUP

FIGURE 2 ATTEMPTED ALTERATIONS OF PARAMETERS

The above were tried individually and in combinations. The predictions were monitored against the data-base. Based on approximately 500 runs of ELESTRES, we selected the combination that gave the best agreement of predictions with the data. Table 3 shows the final forms of the equations for the above parameters. The original value of diffusivity 'D' was multiplied by 2, and is shown in Figure 3. The empirical constants kept the parameters within the ranges of the experimental measurements noted earlier. The next section discusses the results in greater detail.

TABLE 3: FORMS OF THE EQUATIONS USED

Parameter	Form of Equation
1. Thermal Conductivity of UO_2	$k = k_{MATPRO-11} - \Delta k$ (W/(m.K)) Δk = TEMFAC x BURNFAC (W/(m.K)) where TEMFAC: describes the decrease of thermal conductivity as a function of local temperature BURNFAC: describes the variation with burnup
2. Diffusity	$D = 46.8 \times 10^{-6} \exp(-34500/T)$ (cm^2/s)
3. Bubble Radius	The bubble radius for interlinkage BRADMAX = A + Bω (μm) where A & B are empirical constants and ω is the burnup (MW.h/kgU)



$D_1 = 23.4E-6 \times EXP(-34500/T)$

$D_2 = 2 \times D_1$

FIGURE 3: DIFFUSIVITY

4.0 RESULTS

In comparing predictions vs. measurements, one relevant aspect is the scatter in the experimental measurements. A precise number for the scatter in fission gas release measurements has not yet been determined for all CANDU fuel, even though scatter is available for individual experiments. For example, bundle GB [22] was irradiated to outer element burnups of $\sim$ 300 MW.h/kg U, at element powers that declined gradually from 73 to 48 kW/m. Fission gas was measured in 8 out of the 18 outer elements. The seven measurements that were considered reliable showed between 22 to 29% release, i.e. an uncertainty of about $\pm$ 3-4% around a mean of 25-26%.

Figure 4a gives the predictions of the new model. It combines all the 98 irradiations. It also contains a 45° line for reference. Points on this line would indicate a perfect match of predictions vs. experiments; thus the distance of a point from this line indicates the level of discrepancy for that irradiation. Although some points are quite far from the line, the vast majority of the points generally lie close enough to the line.

Figure 4b shows the trend of the deviations (= predictions - measurements) as a function of burnup. The model does not contain any obviously large bias towards any burnup range.

Least squares regression analyses were done on the data by correlating the predictions to the measurements using an equation of the form $y = mx$. Table 4 summarizes the resulting statistics. Figure 4c shows the slope, m, in the four groups of burnups, for the new model as well as for the original model. For all the 98 irradiations combined, the new model has a slope of 1.02, i.e., on average, the new model predicts the measurements to within $\sim$ 2%. Figure 4c also shows that the distribution of the slope with burnup has also improved considerably compared to the original, although further improvements in the slope as well as in the standard deviation are perhaps possible via more sophisticated models.

Figure 4d shows the mean deviations in the four groups of burnups. Again, there is a significant improvement over the original version. The new model has an overall mean deviation of 1.3 percentage points with a standard deviation of 5.8%.

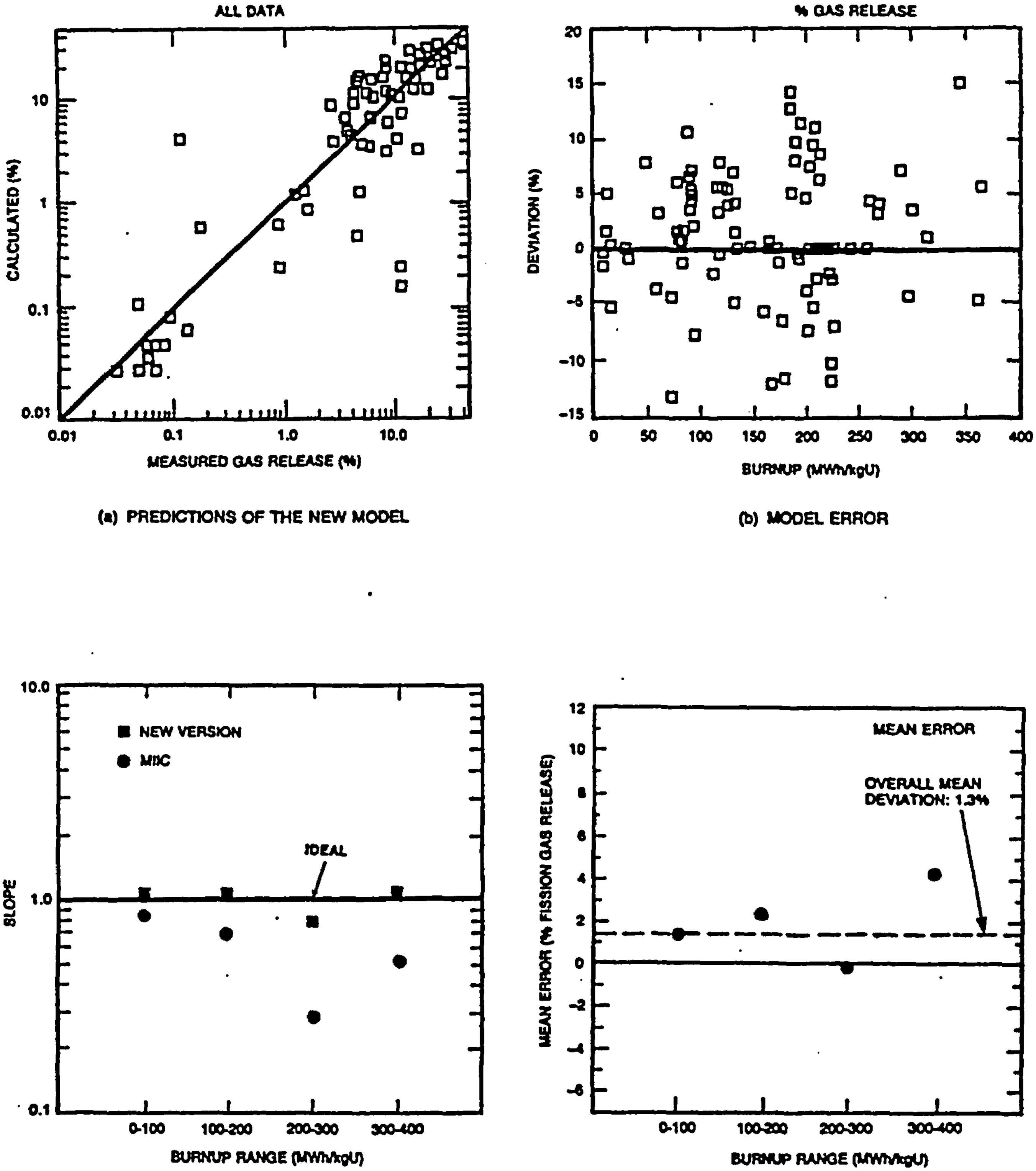
TABLE 4: STATISTICS FROM REGRESSION ANALYSES OF ELESTRES CALCULATIONS VERSUS MEASURED GAS RELEASE

- Based on measurements from 98 irradiations
- Regression Model: y = mx

BURNUP RANGE (MWh/kgU)	PARAMETERS	ELESTRES	
		M11C	NEW VERSION
ALL DATA	SLOPE m	0.6856	1.0200
	STANDARD ERROR OF SLOPE	0.0307	0.0359
	MEAN DEVIATION*	NOT AVAILABLE	1.3228 %
	STANDARD DEVIATION OF MEAN ERROR**	NOT AVAILABLE	5.8356 %
	CORRELATION COEFFICIENT (R^2)	0.6856	0.7235
	NUMBER OF OBSERVATIONS	98	98
	DEGREES OF FREEDOM	97	97

* Mean Deviation $= [\sum_{i=1}^{N} (\text{Predicted} - \text{Measured})_i]/N$

** Standard Deviation $= \sum_{i=1}^{N} \{ [(\text{Predicted} - \text{Measured})_i - \text{Mean}]^2/(N-1)\}^{1/2}$



(a) PREDICTIONS OF THE NEW MODEL

(b) MODEL ERROR

(c) SLOPE BASED ON BURNUP GROUP

(d) MEAN DEVIATIONS BASED ON BURNUP GROUP

FIGURE 4 COMPARISON WITH EXPERIMENTAL DATA

5.0 CONCLUSIONS

1. The data used in this study (98 irradiations) suggest a gradual, rather than abrupt, increase in fission gas release rates with burnup. This conclusion applies to burnups up to 300 MWh/kgU.

2. Modifications have been determined for the following components of the fission gas release model of ELESTRES:

   - thermal conductivity;
   - diffusivity; and
   - density of intergranular bubbles.

   In each case, they are consistent with experimental data.

3. The new model improves the predictions of the ELESTRES code for fission gas release when compared against measurements from 98 irradiations in research as well as in commercial reactors. The data-base covers burnups to 362 MWh/kgU and powers to 110 kW/m. The predictions are now unbiassed with respect to burnup. On average, the predictions now differ from the measurements by 1.3%. For reference, the experimental scatter in a typical irradiation is $\pm$ 3-4%.

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