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Improvements to the Fission Gas Model LAKU and its Coupling to the Pin Behaviour Model URANUS

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Abstract:

The modeling of the fission gas code LAKU has been extended to describe the following effects: The resolution of fission gas from closed pores at the grain edges during steady state irradiation; the separation of grains at the boundaries during transients, caused by either fractures due to the overpressurization of grain face bubbles or by the flooding of the grain faces by released intragranular bubbles; transient grain growth; and, for frothing molten fuel, the time dependent release of the resolved gas. The improvements were performed on both the stand-alone version and URANUS-LAKU. The coupling of the two models URANUS and LAKU is described in some detail, and first results from the coupled code are presented.

Verbesserungen des Spaltgasmodells LAKU und seine Kopplung an das Modell URANUS für Brennstabverhalten

Zusammenfassung:

Das Spaltgasmodell LAKU wurde um die Beschreibung folgender Effekte erweitert: Die Wiederauflösung von Spaltgas in geschlossenen Brennstoffporen unter Betriebsbedingungen; Aufbrechen oder Auftrennen des Brennstoffs längs der Korngrenzen während Transienten, entweder durch Rißbildung aufgrund eines Überdruckes in den Korngrenzenblasen oder durch Übersättigung der Kornoberflächen mit freigesetzten intragranularen Blasen; Kornwachstum bei Transienten; und, für schäumenden geschmolzenen Brennstoff, die zeitabhängige Freisetzung des gelösten Gases. Die Änderungen wurden sowohl an der unabhängigen wie an der an URANUS gekoppelten Version vorgenommen. Die Kopplung von URANUS und LAKU wird im Detail beschrieben und erste Ergebnisse des resultierenden Codes werden gezeigt.

Contents:

1. Introduction

The fission gas model LAKU has been described in detail in 1985 and before /1,2/. Recent work on the model concerned mainly the coupling to the pin behaviour model URANUS /3/. There were, however, some improvements to the LAKU model itself, which were prompted by the simulation of experimental results. The changes of modeling are presented in the first part of the report, together with their effect on the calculational results. The second part deals with the development of the fast version of LAKU, the details of its coupling to URANUS, and the difficulties encountered in some specific cases, which are caused by the interaction of the two models. First results from URANUS-LAKU are presented.

2. Improvements of modeling

A short description of the fission gas model must be given for a better understanding of the following. Fission gas is generated as intragranular gas resolved in the fuel matrix, where it diffuses. It may precipitate into intragranular gas bubbles and, by resolution due to the interaction with energetic fission products, be transfered back into the fuel matrix. There are several mechanisms for releasing the gas to the grain faces or into the pores (fabricated or formed during irradiation): Atomic gas diffusion at low temperatures; grain boundary sweeping - due to the moving grain boundary of growing grains -, sweeping by migrating pores and migration of the intragranular bubbles at higher temperatures. Gas at the grain faces is mostly collected in lenticular grain face bubbles, though a component of gas resolved in the grain boundary is taken into account. Precipitation and resolution are treated analogous to the intragranular case, and gas release to the pores may be by the diffusion of the resolved gas, migration of the grain face bubbles or their interlinkage. Intragranular and grain face bubbles are described with one bubble class each, with an average radius and gas content. Both kinds are assumed to be at equilibrium volume during irradiation, and their density is calculated with simple equations. During a transient, bubble coalescence, overpressure and time-dependent volume equilibration are treated explicitely for both kinds of bubbles.

The pores are modeled as spheres with protruding channels, that grow with time, interlink and form a network allowing gas release. This interlinkage is normally stable, but a model for the formation of a temporary network of tunnels, that collapses after venting, is provided for the case of bigger temperature increases during irradiation and for transients.

A model for gas behaviour in molten fuel during transients is included. It treats three classes of bubbles (the former intragranular bubbles, grain face bubbles and pores) and the resolved gas; precipitation and resolution are modeled, bubbles move due to Brownian motion (small ones) and buoyancy (bigger ones) and may coalesce.

2.1 Resolution of fission gas from closed pores

This improvement to the modeling is the only one concerning the irradiation part of LAKU. It was prompted by the fact that the model gave somewhat high fractions of intergranular gas, especially at low irradiation temperatures. There are very large uncertainties to the measurements of this value, but it is believed not to exceed 20% for temperatures below 1000°C. Therefore, the modeling was checked for simplifications that might unduly enhance the intergranular component.

Up to now, fission gas that has been released to the pores was assumed to remain there until interlinkage and venting. The gas resolution due to the interaction with energetic fission fragments was modeled only for the smaller intragranular and grain face bubbles, whereas resolution from the pores was neglected. There is, however, a quite large amount of gas residing in the pores before venting.First estimates showed, that resolution from these pores may act to reintroduce an appreciable amount of gas into the grain. Therefore it was decided to include the effect in the model.

A very simple estimate of the effect is used for this. The resolution from the pores, R_p, is approximated as

$$R_p = \eta_{eff} \cdot g_p \cdot 1/4$$

g_p is the amount of gas residing in closed pores; η_{eff} is the effective resolution probability; the factor 1/4 takes into account, in a very crude manner, the fact that part of the resolution takes place near grain edges, and that part of the resolved gas is not knocked very deep into the lattice: Both of these effects ensure, that an appreciate part of the gas is directly drained back into the pores. The effective resolution probably is

$$\eta_{eff} = \eta \cdot 10^{-7}/r_p$$

where η is the resolution probability for very small pores, r_p is the pore radius and 10^{-7} is the width of the gas layer, from which resolution can take place. Thus the term

$$R_p = 1/4\ g_p\, \eta \cdot 10^{-7}/r_p$$

describes the source of intragranular gas due to resolution from the pores; it is to be added to the term for gas creation by fission.

Some calculations were performed in order to assess the effect of this change in modeling. The first example concerns the gas release and the intergranular gas fraction of a pin irradiated at low temperatures (mean temperature 1000°C; 1300°C at the center, 650°C at the surface of the fuel). The calculation was performed with the stand-alone version of LAKU, with constant irradiation temperatures. Fig. 1 shows, that the gas release as a function of burnup is not very much affected by the change in modeling; the maximum reduction is about 5% at burnups between 3 and 7%. The effect on the intergranular gas fraction, however, is quite pronounced, as shown in fig. 2.

The second example concerns the post-irradiation state of the CABRI-Rig 1 pins /4/, which were irradiated in the PHENIX-reactor to a burnup of nearly 1% at a linear rating of 430 W/cm, i.e. at high temperatures. The calculation was performed with URANUS-LAKU, and thus the feedback of the change in modeling on gas release and gas driven swelling and hence on pin temperatures was accounted for. It turned out, that the change in the temperatures is not pronounced and not always in the same direction. The total gas release is only marginally reduced, from 45.1% to 44.4%. Fig. 3 shows the axial distribution of the intergranular gas fraction. It is generally higher than in the first example because the grain size is smaller, leading to a higher surface / volume ratio for the grain; this, in turn, favours higher intergranular gas fractions in the LAKU-model. The intergranular gas fraction averaged over the whole pin goes down from 41.9% to 37.5%; this is probably still somewhat high.

2.2. Grain growth during transients

The sweeping effects of grain growth were already modeled in the steady state part of LAKU, but were neglected during transients. One can easily estimate, however, that the grain growth rates measured under steady state conditions lead to a non-negligible sweeping effect for transients at high temperatures with a duration of seconds. The effect was therefore included in the model for both code versions. The main difficulty of this code modification is the modeling of the combination of intragranular gas release by biased bubble migration and by grain growth sweeping, whereas the grain growth function itself is relatively simple and may easily be changed.

The growth formula employed at the moment is the same as for steady state growth:

$$a^4(t) = a^4(0) + t \cdot c_1 \cdot \exp(-c_2/T)$$
$$a(t) \leqq c_3 \cdot \exp(-c_4/T)$$

(a grain radius; t time; T temperature; c_1 - c_4 constants)

It describes a temperature dependent growth with a temperature dependent upper limit. In addition, the growth is slowed down, when the fraction of the grain face covered by bubbles exceeds .5, and it is totally stopped, when this value exceeds .8.

A simulation of the experiment FGR-41 /5,6/ has been used to assess the effect of this addition to the model. The experiment has been performed out-of-pile. A small slug of fuel was submitted to a transient with the fuel temperatures increasing by up to 200°C/s, and the time dependent fission gas release was measured. Fig. 4 shows the results of a simulation with the stand-alone version of LAKU, with and without the transient grain growth model, and in addition with a variant of the model with grain growth uninhibited by the grain surface bubbles. A noticeable effect occurs only in this last case, whereas there is practically no difference between the old model and the more realistic one with conditional growth. It must be stressed however that this result is far from general, but may be subject to the transient chosen and the parameters of the model.

2.3. Model for grain boundary separation

A grain boundary separation may be caused by either the saturation of the grain surfaces by released intragranular bubbles or by a stress-induced fracture. A subroutine has been added to both versions of LAKU, which determines, whether grain boundary separation takes place, and the model has been changed to take into account the consequences of such a separation.

The saturation by grain surface bubbles is determined by simply examinig the fraction of the grain surface covered by bubbles. This value is kept at or below a maximum value, currently 80%, under steady state conditions; at this value, total interlinkage is assumed to occur, and all gas arriving at the surface later on is drained directly into the pores. The same assumption was used under transient conditions, but this has now been dropped and the covered fraction is allowed to go up to 100%. Interlinkage in zones that do not have interlinkage at the start of the transient is assumed to occur at 80% covering, as before. The grain boundary separation is assumed to occur at a 10% higher value, i.e. at 88%.

The model of Worledge /7/ forms the basis of the model for stress-induced grain boundary separation. He has formulated the following 5 conditions, which have to be fulfilled simultaneously for a stress-induced fracture to occur:

- stress condition: The stress induced by the exess pressure in the grain surface bubble must exceed the yield stress of the material

  $P_{ex} \geqq \sigma_y/f$

  (p_{ex} excess pressure; σ_y yield stress; f stress concentration factor at the tip of the crack opening up the grain boundary)

- Differential energy criterion: The energy required for advancing the crack must be supplied by the bubble gas expanding into the additional volume

  $P_g \geqq 2Ws/\delta$

  (p_g pressure in the grain surface bubble; W_s free surface energy; δ crack width)

- Pressure increasing: The bubble pressure, as determined in a bubble dynamics calculation that ignores cracks, should be increasing in order to ensure a continuous crack advance:

$P_g > 0$

- Total energy criterion: The total energy required to form the surface up to crack interlinkage with neighboring cracks must be supplied by the expanding gas.

$E_{gas} > E_{surface}$

This criterion reduces to the second one for cases, in which the bubble volume is much greater than the volume of the crack.

- Mass transfer condition: Crack healing due to the vacancy diffusion to the bubble should be slower than the stress-induced crack advance:

$|V_{sp}| < V \cdot P_g/P_g$

(V_{sp}: volume flux from crack tip to bubble; V bubble volume)

These five criteria are currently used in LAKU. If they are fulfilled, grain boundary separation is assumed to take place, the grain face gas is released to the pores, and the grain face bubbles are assumed not to migrate and coalesce any more; their volume increases only by the volume of the released intragranular bubbles, and grain growth is stopped.

The experiment V83 /8/ has been used as a first example for testing these criteria. This in-pile experiment consists of a fuel slug being subjected to an energetic pulse of short (1ms) duration; the energy deposition is strongly space-dependent, due to a pronounced neutron self-shielding, which leads to a large temperature gradient in the fuel. The hottest parts of the fuel experienced melting. Extensive grain boundary micro-cracking has been observed in the cooler parts of the sample.

LAKU in its present version has not been able to reproduce the cracking, since not all of the criteria were fulfilled. There has been speculation /9/, that the big temperature gradient in this experiment enhances the probability for cracking; possibly the criteria should be modified to include this effect. In addition, further experiments should be simulated with the code in order to verify the cracking model. The FD4 experiment series is particularly suited for this, since a solid state fuel dispersal has been observed in some of these experiments (e.g. FD4.3 /10/) and the time of dispersal and the fuel energetics are well known.

2.4 Gas release from frothing fuel

No attempt was made in LAKU up to now to model the gas release from frothing fuel. When frothing occured, i.e. when the volume of the gas in the molten or melting fuel exceeded that of the fuel, the calculation (for that zone) was simply stopped and all gas assumed to be released. The frothing condition may however be fulfilled right at the onset of melting, and if this happens in an unrestructured fuel zone during a fast transient, a large part of the gas may still be resolved in the fuel matrix. This gas will not be released immediately but only after the time it needs to diffuse to the next free surface.

The revised model is not stopped after the onset of frothing any more. All gas contained in bubbles may expand freely, and the behaviour of the bigger bubbles is not simulated, as before. The calculation is continued, however, to treat the diffusion of the resolved gas to the group of the smallest (and most abundant) bubbles, the former intragranular ones, and is stopped only when all gas has reached the bubbles.

The simulation of V83 cited above has been used to test the effect of this correction. Fig. 5 shows the time dependent gas release from a cross section of the slug for the old and the corrected model, employing a diffusion coefficient for the resolved gas in the molten fuel of 10^{-7} cm^2/s. Evidently the gas release is markedly delayed by the diffusion process. A quantitative comparison with the measured gas release is not possible, since the simulation treats only one cross section of the sample, and the temperature profile in this case depends strongly on the axial position. The measured gas release is, however, somewhat faster than the results of the new model. Probably the diffusion coefficient should be augmented to $3\text{-}10 \cdot 10^{-7}$ cm^2/s. This would put it nearer the range of known estimates, 10^{-6} - 10^{-5} cm^2/s.

3. URANUS-LAKU

The coupling of LAKU to the pin behaviour model URANUS consists of three steps:

- development of a somewhat simplified fast version of LAKU;
- coupling itself, i.e. the substitution of the LAKU- input/output with a transfer of data from and to URANUS;
- testing of the resulting code.

Work has currently progressed to the third step, though the first one may need more perfection.

3.1 Development of the fast version of LAKU

The fast version is very similar to the stand-alone code, since the physical model has been kept intact. Below the most important approximations in the fast version, as compared to the slow one, are listed:

- The function describing the dependence of the intragranular bubble velocity on the bubble radius is simplified.
- The error margin for solving the differential equations numerically is augmented by one order of magnitude.
- The high temperature enhancement of the creep process is deleted.
- The numerical treatment of the steady state development of the gas distribution is greatly simplified by making extensive use of asymptotic balance conditions.
- The model for the transient release of gas from the open porosity (streaming through the interlinked pores to the surface of the fuel) is deleted.
- The double precision routines are replaced by single precision ones.

The fraction of computer time used by LAKU in productions runs of URANUS-LAKU stands currently at 50%. This value should be somewhat decreased, especially for the transient part.

3.2 Coupling of URANUS and LAKU

Apart from purely organisational changes, the main task of coupling codes consists in establishing the data transfer among the models. Thus the external input of LAKU has to be replaced by an internal transfer of URANUS - results, and some of the results of LAKU have to be transferred back to URANUS. This defines then the interaction of the two physical models. The data transfer, as it stands at present, is specified below:

- URANUS results transferred to LAKU.
  Fabrication data (fuel porosity and grain size, pin geometry); length of the individual time steps; organisational data (steady state or transient irradiation?, first calculation of a time step or iteration?); time dependent results (ambient pressure in the pin, axial distribution of contact pressure, axial and radial distribution of gas creation rates, fuel temperatures, melting temperatures and porosity). The grain sizes are calculated independently by LAKU, but with the growth law employed by URANUS.

- Material constants employed by LAKU.
  They can be changed by input in the stand-alone version. However, they are normally kept constant, since they are fitted together with the modeling to reproduce sufficiently well a spectrum of steady state and transient experiments, and are changed only together with the model. Constant values are therefore used in the coupled code.

- LAKU results transferred to URANUS.
  The following time dependent axial and radial distributions are transferred back: The quantities of intra- and intergranular gas, gas driven swelling and the strain caused by fission products, i.e. including non-gaseous products.

Some more data may have to be transferred to LAKU in the future, namely:

- Information on whether the reactor is a thermal or a fast (or intermediate) one.
  The gas creation rates are used in LAKU to infer the fission rates; these, in turn,

are used to compute some material constants. This process involves some error if the neutron spectrum is not taken into account, since the creation rates vary by some 10-20% from fast to thermal. Currently the material constants are changed for each simulation of a new reactor to arrive at the correct data. This process can be avoided by using the information that exists in URANUS.

- Fuel enthalpy during melting.

The present model switches to the model for gas in molten fuel, when the temperature reaches the melting point. A more realistic model must take into account, that the fuel does not behave as a liquid throughout when it reaches the solidus,but does so only at some higher enthalpy. Thus a better melting model will need the fuel enthalpy in addition to its temperature.

The coupled model works in the following way: A time step length is chosen by URANUS using a sophisticated routine. Then URANUS does the necessary thermodynamics calculations, calling LAKU in their course; it ends by comparing the relevant data with their estimates and deciding on whether an iteration is necessary. The time step length remains unchanged for the iterations, but the length of successive time steps is adapted to the physics.

The code has been tested and is working, but problems are encountered in some special situations. These problems do not pertain to one or the other code, but stem from the interaction of the two. Mathematically speaking, the iteration does not converge, when the numerical problem posed by the combination of the two models becomes highly nonlinear. This happens in two cases:

- Closing of the gap between fuel and cladding during steady state operation

The problem results from the (somewhat unphysical) assumption in LAKU, that intra- and intergranular fission gas bubbles instantaneously adopt their equilibrium volume during irradiation. With an open gap, fuel temperatures are high and the contact pressure is zero; thus the bubbles tend to be big and contribute to gap closure at some point during the irradiation. At this point, temperatures drop noticeably and the contact pressure rises sharply. An iteration becomes necessary, now with the lower temperatures and the new contact pressure; the LAKU model responds to both with somewhat smaller bubbles, i.e. smaller fuel expansion, and the gap reopens. This leads often to an oscillation of the successive iterations between closed and open gap. At present, time step lengths are sharply reduced when the gap nears closure, and the increase in contact pressure is limited for the use in LAKU (thus it may happen that the "real" contact pressure provided by URANUS is used in LAKU only after a few time steps). In this way, the calculations mostly proceed successfully beyond gap closure, but divergent cases are nevertheless encountered occasionally.

- Onset of melting in confined gassy fuel.
This leads to a sharp increase in gas driven swelling, that should be countered by a sharp rise in pressure from URANUS. Mostly, however, the numerics diverge or oscillate. The problem has not yet been tackled, but is linked again to an unphysical assumption in LAKU: The instantaneous switching to the melting model. A model providing for a gradual outset of melting (see above) may improve the problem.

3.3 Calculation of the irradiation of the CABRI-Rig 1 in PHENIX

An example for a successful simulation with URANUS-LAKU is the recalculation of the irradiation of the CABRI-Rig 1 pins in PHENIX mentioned in 2.1. This calculation proceeded without complications, because the gap does not close in the short irradiation span, up to nearly 1 at. %. The overall gas release is 44.4% calculated vs. 40.0 ± 4.5% measured. Fig. 6 shows a comparison of the calculated and measured axial gas distribution. Two measured results of the AI-1-experiment are included, because the energy release at the ends of the pin was very small in this experiment, and the gas content is therefore expected to remain unchanged. The agreement between experiment and calculation is very good except for a small deviation at the lower end of the pin.

4. Conclusions

The fission gas model LAKU has been improved in some details, mostly concerning the modeling of transients. The coupled code URANUS-LAKU is operative and first results are very satisfactory. Numerical problems tend to turn up, however, for two special situations, gap closure during irradiation and fuel melting. The following work is planned in the immediate future:

- verification of the fragmentation model;

- improvement and verification of the model for melting fuel;

- coupling to the newest version of URANUS (TRANSURANUS);

- improvement of the iteration technique and / or the physical models leading to instabilities;

- acceleration of the transient part of LAKU.

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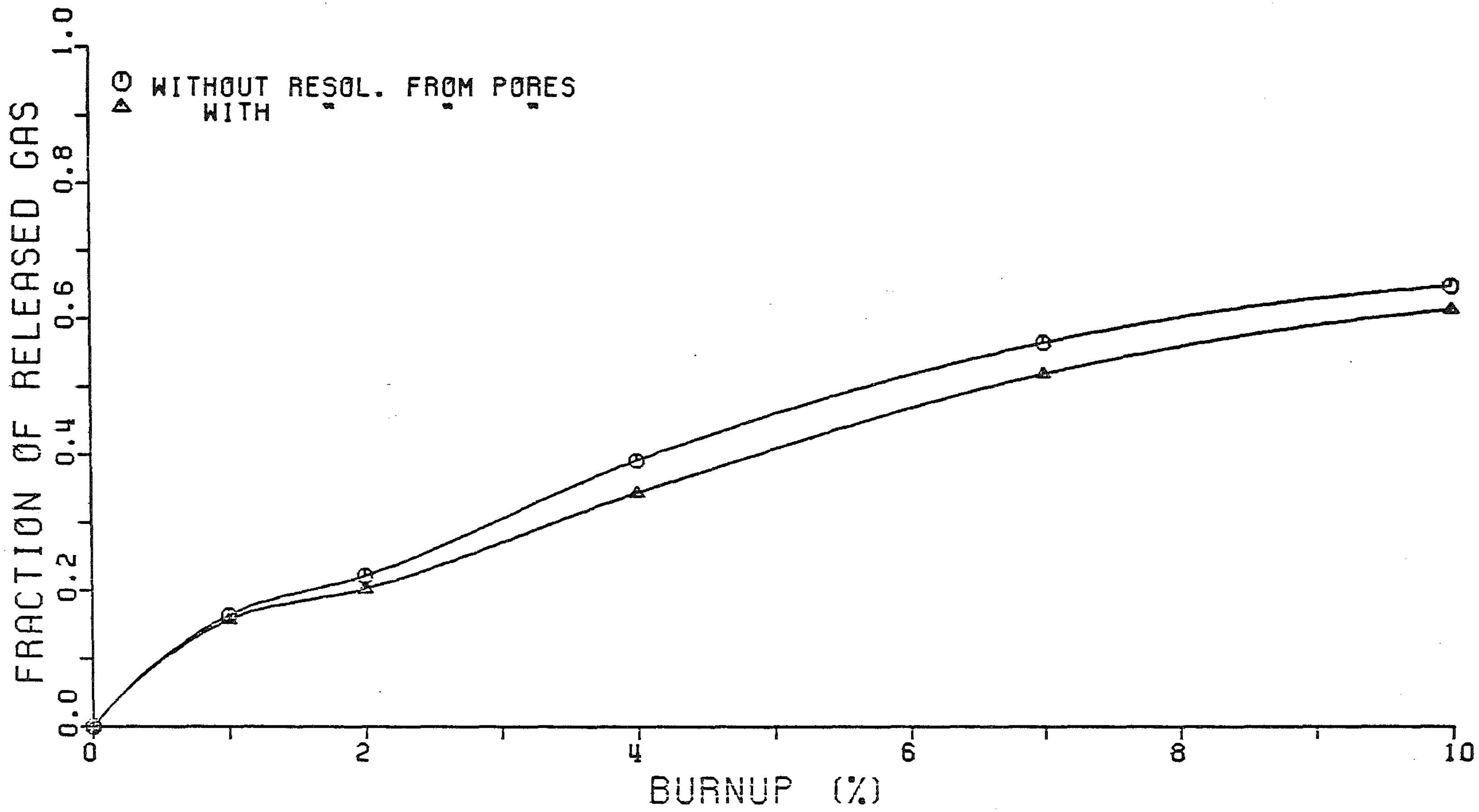

FIG. 1: BURNUP DEPENDENT GAS RELEASE FOR A PIN IRRADIATED AT LOW TEMPERATURE

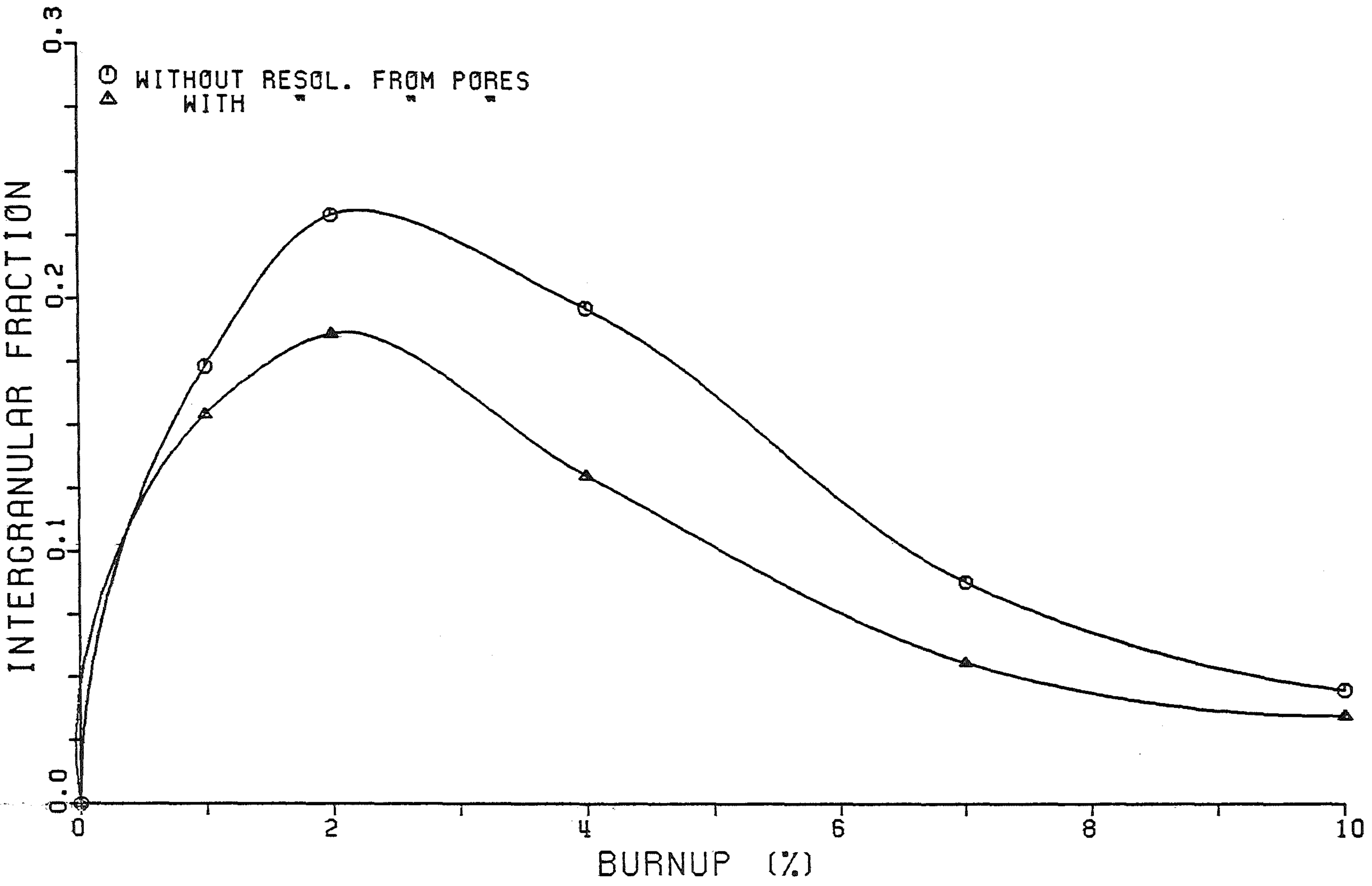

FIG. 2: BURNUP DEPENDENT INTERGRANULAR GAS FRACTION FOR A PIN IRRADIATED AT LOW TEMPERATURE

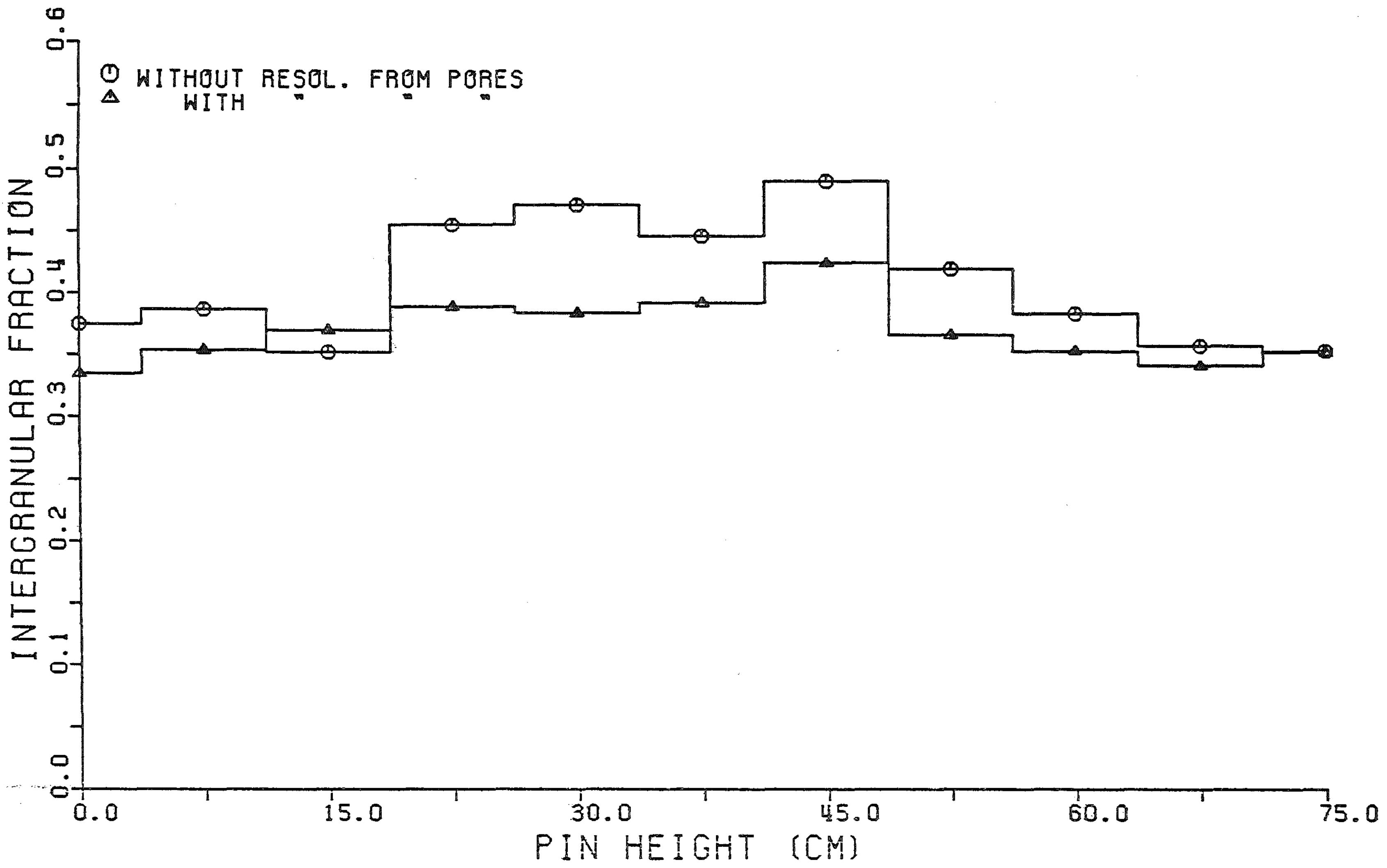

FIG. 3: AXIAL DISTRIBUTION OF THE INTERGRANULAR GAS FRACTION
IN THE CABRI RIG1 PINS AFTER STEADY STATE IRRADIATION

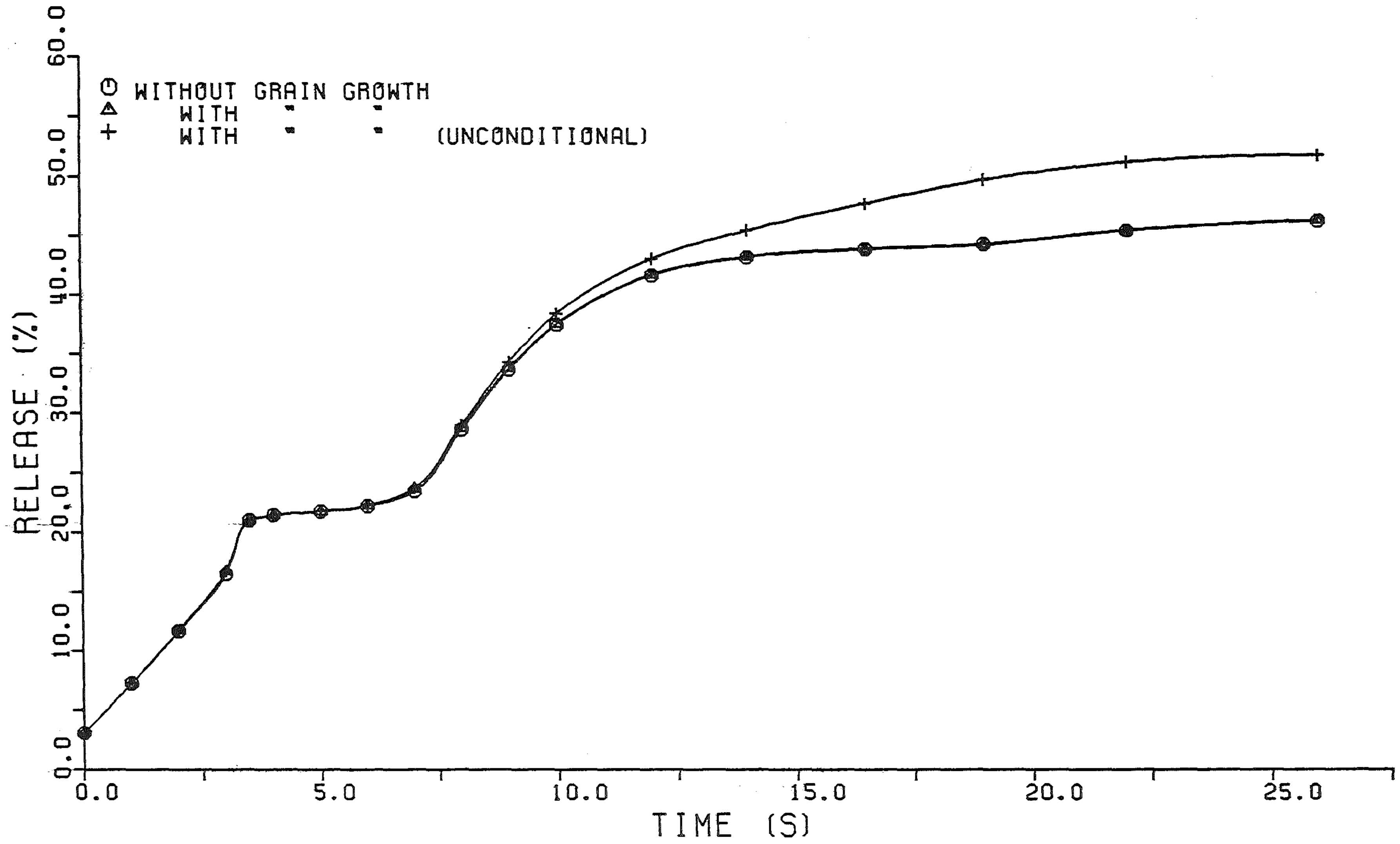

FIG. 4: GAS RELEASE FØR HEDL-TRANSIENT FGR-41.
LAKU-RESULTS WITH AND WITHOUT TRANSIENT GRAIN GROWTH.

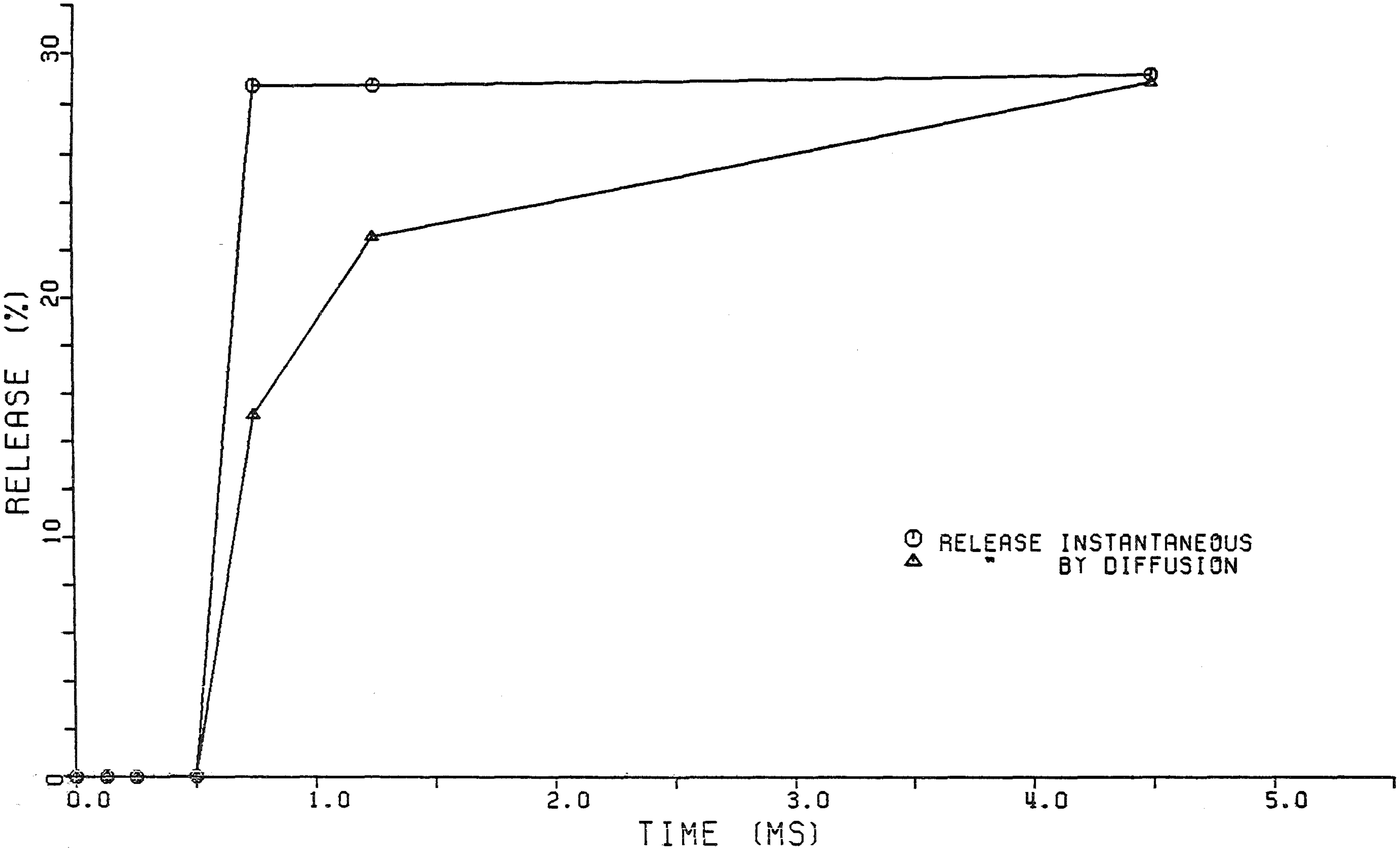

FIG. 5: GAS RELEASE FOR VIPER-EXPERIMENT V83; DIFFERENT MODELS FOR THE RELEASE OF THE ATOMIC GAS FROM FROTHING FUEL.

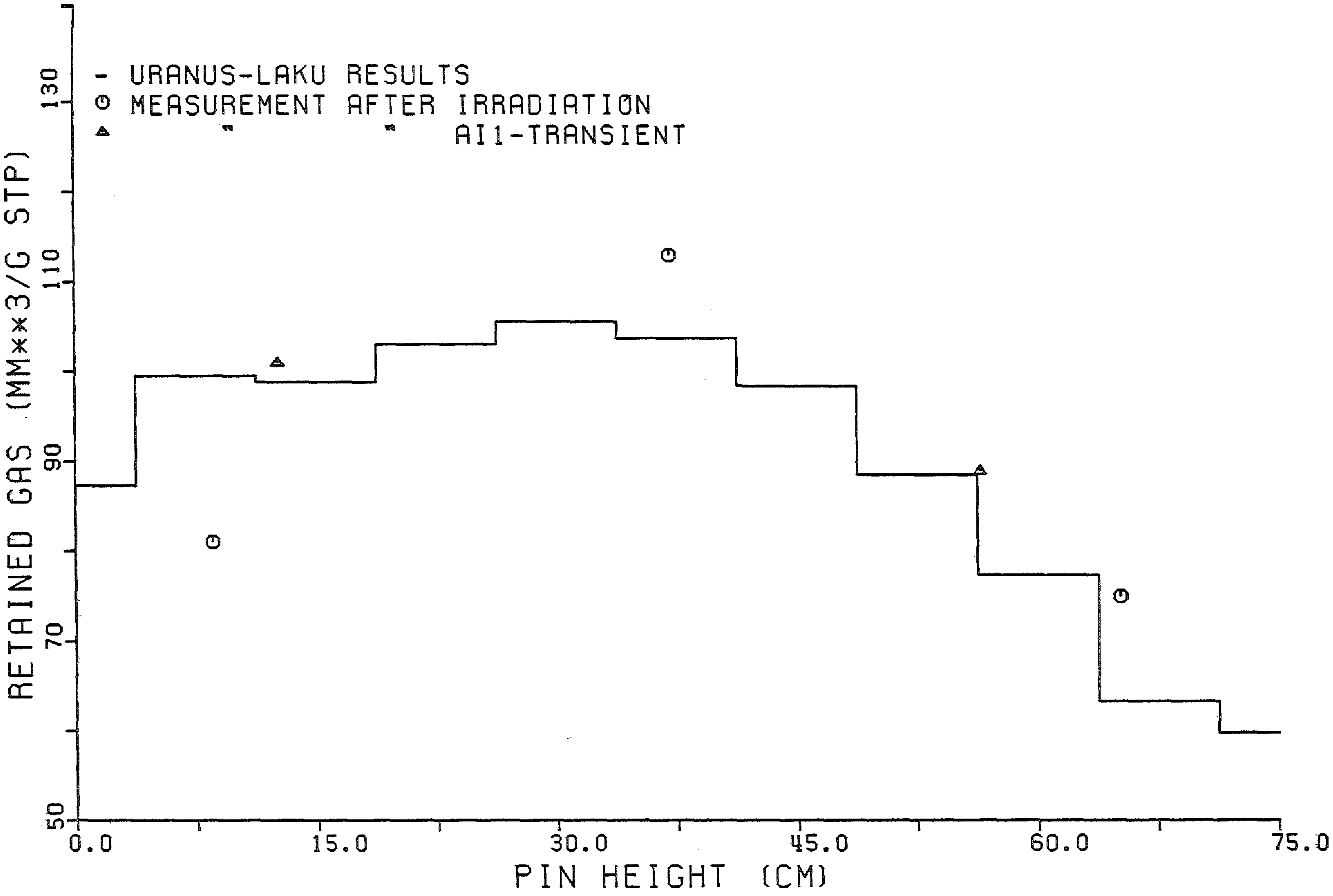

FIG. 6: AXIAL DISTRIBUTION OF THE RETAINED GAS IN THE CABRI RIG1 PINS AFTER STEADY STATE IRRADIATION