Analytical Study of Criticality Experiments of Organic and Light Water Moderated Mixed Oxide Fuel Pin Arrays

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有機溶液及び軽水減速FTR燃料炉心の臨界安全実験解析

相原永史[*1]，福村信男[*1]，角谷浩享[*2]
小山智造[*3]，M.J.Haire[*4]

要　　旨

日米共同臨界実験の中の一項目として，有機溶液及び軽水減速材とMOX燃料ピンの格子からなる炉心の臨界実験が行なわれた。この実験結果に対してベンチマーク解析を実施し，精度評価を行った。

解析コードはATRの解析を通じて高精度の実績を得ているWIMS-CITATION結合システムを用いた。解析手法はエネルギー6群，3次元形状モデルを用い，減速材中の水素の散乱核を計算するために，有機溶液及び軽水共通にネルキンモデルを用いた。

解析の結果，軽水減速材炉心はKeff=1に対して，0.2%ΔK以内の解析精度で良い一致を示したが，これに比べて有機溶液減速材炉心は，格子ピッチが大きくなるに従い，僅かな過小評価を示すことが分った。これは，格子ピッチが大きくなると減速材の占める割合が増大することに伴って，有機溶液と軽水の化学結合モデルの違いによる影響が現れてくるものと考えられる。

今後，有機溶液減速材特有の中性子散乱核モデルを作成して，軽水減速材の場合と同等の解析精度を確立させる必要がある。

[*1] 大洗工学センター，実験炉部，臨界工学試験室

[*2] 大洗工学センター，臨界工学試験室嘱託（センチュリ・リサーチ・センター（株））

[*3] 再処理部，計画課

[*4] オークリッジ国立研究所，燃料サイクル部

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Analytical Study of Criticality Experiments of Organic and Light Water Moderated Mixed Oxide Fuel Pin Arrays

N. Aihara[*1], N. Fukumura[*1], H. Kadotani[*2]
T. Koyama[*3] and M.J. Haire[*4]

Abstract

As part of a joint criticality data development between the Power Reactor and Nuclear Fuel Development Corporation (PNC) of Japan and the United State Department of Energy (USDOE), critical experiments have been conducted with organic moderated Fast Test Reactor (FTR) mixed oxide fuel pin arrays. The neutronic characteristics of an organic moderator can be examined by comparing the results of these experiments with the results of the same type of experiments performed with light water moderated system. In recent experiments performed at the Battelle Pacific Northwest Laboratories Critical Mass Laboratory, five distances of the lattice pitches were utilized which span from soft to hard neutron energy spectra.

Results obtained by benchmark analyses of these experiments are discussed in this paper. The benchmark analysis was performed using the coupled WIMS-CITATION computer code whose accuracy has been demonstrated by applying them to heavy water reactor (ATR) with cluster type fuel. The scattering kernel used in the calculation for hydrogen atoms in light water was based on

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[*1] Criticality Engineering Section, Experimental Reactor Division O-arai Engineering Center, PNC.
[*2] Criticality Engineering Section, (Non-regular staff, on leave from Century Research Center Corp.).
[*3] Planning Section, Reprocessing Division, Head Office, PNC.
[*4] Fuel Recycle Division, Oak Ridge National Laboratory.

Nelkin's model. This same scattering kernel was used for hydrogen atoms in the organic moderator.

The agreement between calculation and experiment (k-eff=1) for the light water moderated cores are fairly good (less than 0.2% Δk/k). On the other hand, k-eff's for the organic moderated core show a tendency to diverge from k-eff=1 gradually, as the lattice pitches become larger; the approximation used to calculate the scattering kernels for the organic moderator becomes less accurate.

It is theorized that small difference in chemical bonding between light water and organic moderator account for the increasing deviation from k-eff=1 for the organic moderators, the larger the fuel pin lattice pitch. Future work would be necessary to establish a more accurate scattering kernel model of the organic moderator to precisely calculate critical conditions for organic moderated fuel pin arrays with large lattice pitches.

Contents

1. Introduction

We summarized part of criticality data obtained from the joint program between the Power Reactor and Nuclear Fuel Development Corporation (PNC) of Japan and the United States Department of Energy (USDOE).[1] The criticality safety experiments which are discussed in the present paper are conducted with organic moderated Fast Test Reactor (FTR) mixed oxide fuel pin arrays. The obtained critical number of fuel pins were compared with the results of same type of experiments performed with light water moderated systems.[2] The obtained experimental results are shown in Table 1.

Five distances of the lattice pitches were utilized which span from soft to hard neutron spectra. These lattice pitches range from 0.761 to 1.935 cm which correspond to a volume ratio of 1.62 to 18.13. In the present analysis, we took the following issues obtained by examining the experimental results.

(1) The number of critical fuel pins are same for the light water moderated and organic moderated systems having the lattice pitch 1.537 cm. These two critical experiments were conducted at the same date which is 1985. Since the moderator was different in each system, we expect the different fuel pin number should be observed. We will try to explain this accidental agreement.

(2) The other experiments were conducted at the different date, namely 6 to 7 years. So, the number of critical fuel pins is different for light water moderated and organic moderated systems due to accumulation of Am-241 or in other words decay of Pu-241.

(3) We have not included in the present analyses the tight pitch system that has the pitch length of 0.761 cm. Because this critical system has the extremely hard spectrum as a thermal reactor. And also, this system was two region core by adding fuel pins having higher enrichment.

Basically, the benchmark analyses have been performed against these light water moderated and organic moderated systems and evaluated accuracy of the theoretical calculation.

2. Experiment

The result of experiments is discussed in this chapter. Figure 1 shows the layout of final subcritical loading arrangement for 1.537 cm lattice pitch core. The detectors for neutron counting are located at three places as shown as data channel 1, 2 and 3. The inverse of the counting rate from those detectors are used to extraporate the critical fuel pin number from the subcritical core.

Figure 2 shows the measured inverse neutron counting rate from three neutron detectors. The horizontal axis is for the loaded fuel pin number and the vertical axis is for the inverse neutron counting rate.

As you can see from this figure the neutrons were measured at the cores loaded with 180 and 195 fuel pins. The inverse of those neutron counting rate are linearly plotted against the loaded fuel pin number and linearly extrapolated to zero inverse neutron count to obtain the critical fuel pin number. This critical fuel pin number was determined by averaging the data from three data channels.

The structure of FTR fuel pin is shown in Figure 3. Besides the MOX fuel region, we took into account in the analysis the thermal shield made of natural uranium and the reflector made of inconel. The content of the fuel is the analytical data for the year of 1976. Therefore, the decay of Am-241 was corrected to the actual date that the experiment was performed. This corrected number density was utilized in the analysis.

The comparison of contents between light water and organic moderator is shown in Figure 4. Needless to say, the light water consists of 100 wt% H_2O molecule, while, the organic moderator consists of 38 wt% tributyl phosphate (TBP) and 62 wt% normal paraffin hydrocarbon (NPH). This NPH is the mixture of five different materials. The atom density of light water is 67% hydrogen and 33% oxygen, which can be compared to that of organic moderator, that is, the content of oxygen is extremely small, 3%, and the content of carbon is 30%. By comparison of molecules which compose light water and the organic moderator, we expect the chemical bonding effect can be observed as the difference of critical fuel pin number in light water moderated and organic moderated cores.

3. Analysis

3.1 Code System

WIMS-CITATION[3,4] coupled code system was utilized for the present analysis. The homogenized cell group constants were produced using WIMS code, which is developed at Winfrith, UK. The neutron transport is treated in the frame work of the collision probability method. The cross section library of WIMS code has 69 group structure. Six broad group constants were generated from this library considering the resonance of various nuclides of plutonium.

The core analysis was calculated with CITATION code which was developed at Oak Ridge National Laboratory. The effective multiplication factor (k-eff) was obtained by three dimensional geometry model in the diffusion approximation. The number of spatial meshes is 25 in X direction, 25 in Y direction and 22 in Z direction for the upper half core. The calculational model of core consists of MOX fuel region, natural uranium shield region, inconel region and reflector. This is the so-called multi layer modelling for three dimensional analysis. The outline of code system utilized in the analysis is shown in Table 2.

3.2 Procedure

The actual procedure of analysis is described in Table 3. The critical fuel pin number in the experiment was determined by extrapolating the inverse of neutron counting rate of the subcritical state. Therefore, the real critical condition was not achieved experimentally, in other words, the final several fuel pins which are necessary to make the system critical were not loaded into the core.

In the present analysis, we proposed the smeared fuel pin model to take into account this last fuel pins. And to justify this smeared fuel pin model we conducted the direct simulation of inverse neutron count experiment. The Neklin's model[5] was used, tentatively, to calculate the scattering kernel for both light water and organic moderators.

The horizontal and vertical plane of three dimensional geometrical model of the calculation is shown in Figure 5. The moderator region was divided into two regions, as shown in the holizontal plane of this figure, to take into account the neutron spectrum change. The vertical regions consist of MOX fuel region, uranium thermal shield region, inconel region and reflector region.

The geometry of critical core used in the analysis is, as shown in Figure 6, composed of two regions, namely the subcritical core which was determined by the experiment, and the added smeared fuel pin region. The smeared fuel pin region is the assumed region which was not loaded in the experiment and the volume equivalent of the fuel pins to be added to make the system critical.

The method used to simulate the inverse neutron count measurement is described in Table 4. The purpose of this analysis is to verify the proposed smeared fuel pin model. We obtained the inverse neutron count curve with calculated thermal neutron flux. Spontaneous fission neutrons from fertile materials were used as the external neutron source.

The points of flux used to obtain the inverse neutron count curve are compared with the experimental data channels in Figure 7. The core of this figure is the light water moderated system having 1.537 cm lattice pitch. Measured points 1, 2 and 3 correspond to calculated points 1, 2 and 3, respectively. Neutron flux for two subcritical cores, which are cores of 15×12 and 15×13 fuel pins, were calculated using spontaneous fission neutron source. And then, the critical fuel pin number was estimated by extrapolating the inverse of thermal neutron flux of two subcritical cores.

An example of the calculated inverse neutron flux is shown in Figure 8. The critical fuel pin number was obtained by averaging zeros of each inverse neutron flux curve of flux points 1, 2 and 3. The theoretical critical fuel pin number thus obtained is larger than the experimental value by 1.4 pin. And this is greater than the experimental error, which is 0.3 pin.

We compared in Table 5 the critical fuel pin number calculated with simulated inverse neutron count method with the smeared fuel pin model method for the light water moderated cores of 0.968 cm and 1.537 cm lattice pitches. The critical fuel pin number obtained by the simulated inverse neutron count method is less than the experiment by 2.6 fuel pins for 0.968 cm pitch core and greater than the experiment by 1.4 fuel pin for 1.537 cm pitch core. These differences correspond to +0.10% Δk and −0.18% Δk, respectively. On the other hand, the error of analysis using the smeared fuel pin model is +0.09% Δk for 0.968 cm pitch core and −0.23% Δk for 1.537 cm pitch core, respectively. Agreement between two calculational models is very good, that is, less than 0.1% Δk. We confirmed that the case of organic moderated cores also obtained same good agreement between two calculational models as the case of light water moderated cores, as shown in Table 6. Therefore, we can conclude that the smeared fuel pin model is applicable to the present experiment. So, we utilized this analytical model throughout the present analysis.

4. Result and Discussion

The results of k-eff of both light water moderated and organic moderated systems are shown in Table 7. We would like to exclude from the present analysis the core having 0.761 cm lattice pitch, because v-m/v-f ratio of the core is extremely small, which means that the neutron spectrum in this core is very hard compared to the other systems. Furthermore, to achieve this core critical, higher enriched fuel pins were added at the peripheral region of the core, which makes this core heterogeneous, while the other core was loaded uniformly.

The calculated k-eff of the light water moderated core agreed well with the experiment. The error of k-eff is less than 0.2% Δk. On the other hand, k-eff of the organic moderated core deviate from k-eff=1 gradually, as the lattice pitch becomes larger.

The calculated k-eff's of both light water and organic moderated systems are shown graphically in Figure 9. As described previously, k-eff of light water moderated cores agreed quite well with the experiment, that is less than 0.2% Δk. The organic moderated core of 0.968 cm lattice pitch shows the same accuracy as the light water moderated core. However, the deviation from k-eff=1 becomes larger, as the distance of lattice becomes larger. This tendency seems to saturate for the core which is larger than 1.537 cm lattice pitch. As the volume ratio of moderator to fuel becomes larger, the error of the analysis becomes larger.

In other words, for the core of 1.537 cm lattice pitch the calculated critical fuel pin number of the light water moderated core agrees well with the experiments, while the critical fuel pin number of the organic moderated core is calculated to be larger than the experiment. This means that the scattering kernel model, that is Nelkin model, is adequate for light water but this model is not applicable to organic moderator. This can be easily understood, because the molecular structure and the characteristics of chemical bonding of organic moderator is quite different from light water.

To obtain the idea of the chemical bonding effect of the criticality calculation, we repeated the same calculation using the free gas model for the calculation of the scattering kernel, as shown in Table 8 and Figure 10. The free gas model is the one extreme example of calculation which neglects entirely chemical bonding effect. We calculated the k-eff's for both light water moderated and organic moderated systems of 0.968 cm and 1.537 cm lattice pitches. The calculated k-eff's are greater than unity by 1.0 to 1.5% Δk compared to the results of Nelkin model.

So, we can conclude, i.e.

(1) The Nelkin model is not applicable to organic solution for the calculation of scattering kernel.

(2) The calculated reactivity depends on the scattering kernel model.

Therefore, we can say that the accidental agreement of the critical fuel pin number is caused by chemical bonding effect of the light water and organic moderator.

5. Conclusion

The conclusions are summarized as follows:

(1) Chemical bonding effect induced accidental agreement of critical fuel pin number between water moderated and organic moderated systems (for 1.537 cm lattice pitch).
   This was concluded by:
   - Disagreement of k-eff calculated for water and organic systems using same scattering kernel model (Nelkin model).
   - Reactivity difference induced by changing kernel models (Nelkin and Free gas model).

(2) Accuracy of analysis is quite good for water moderated systems (<0.2% for k-eff). Inadequacy using Nelkin kernel in organic system is increased for systems having larger lattice pitch.

(3) Smeared fuel pin model was shown to be accurate for analysis of the present experiment by direct simulation of inverse neutron count

measurement.

Future work would be necessary to establish a more accurate scattering kernel model of the organic moderator to precisely calculate critical conditions for organic moderated fuel pin arrays with large lattice pitches.

6. References

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Table 1. Experimental Criticality Data – FTR Fuel Pins Moderated and Fully Reflected by Either Water or an Organic Solution of TBP–NPH

	Water Moderator (b)			Organic Moderator (b)		
Lattice Spacing [a][b] (cm)	Experiment Number	Date	Critical Number of Fuel Pins	Experiment Number	Date	Critical Number of Fuel Pins
0.761±0.001	067	7-17-85	1046.9±0.2	065	6-5-85	1054.8±0.2
0.968±0.001	021	11-3-78	571.9±0.2	063	5-21-85	599.2±0.8
1.242±0.001	043	1-9-79	293.9±0.1	062	5-15-85	301.8±0.2
1.537±0.001	068R	7-25-85	199.7±0.3	061	5-14-85	199.5±0.3
1.935±0.002	032	12-14-78	165.1±0.4	060	5-8-85	165.3±0.1

(a) Center-to-center spacing between fuel pins in a square pattern

(b) Error limits are one standard deviations

Table 2. Code System Utilized in the Analysis

Code System

WIMS—CITATION Coupling System

Lattice

Collision Probability
69 Groups Library
6 Groups Homogeneous Lattice Constant

Core

Diffusion Theory
3 Dimensional Geometry
Fine Mesh : 25 × 25 × 22 (Upper Half Core)
Multi Layers : MOX, UO_2, Inconel, Reflector

Table 3. Procedure

- Establishment of Critical Core Configuration
  - Smeared Fuel Pin Model
  - Simulation of Inverse Neutron Count Measurement
- Scattering Cross Section of Hydrogen
  - Water : Nelkin Model
  - Organic : Nelkin Model

Table 4. Simulation of Inverse Neutron Count Measurement

Verification of Smeared Fuel Pin Model

Inverse Neutron Count Simulation Using Thermal Neutron Flux

Source : Spontaneous Fission Neutrons of Fertile Material

Table 5. Verification of Smeared Fuel Pin Model (Water Moderator)

Lattice Pitch (cm)		Simulated Inverse Neutron Count		Smeared Fuel Pin Model	
		Critical Number of Fuel Pins	%ΔK*1	K_{eff}	%ΔK*1
0.968	Experiment	571.9	+0.10	1.0	+0.09
	Calculation	569.3		1.00089	
1.537	Experiment	199.7	−0.18	1.0	−0.23
	Calculation	201.1		0.99770	

*1 Calculation - Experiment

Table 6. Verification of Smeared Fuel Pin Model (Organic Moderator)

Lattice Pitch (cm)		Simulated Inverse Neutron Counts		Smeared Fuel Pins Model	
		Critical Number of Fuel Pins	%ΔK[*1]	K_{eff}	%ΔK[*1]
0.968	Experiment	599.2	+0.03	1.0	+0.02
	Calculation	598.3		1.00019	
1.537	Experiment	199.5	–0.71	1.0	–0.76
	Calculation	205.0		0.99237	

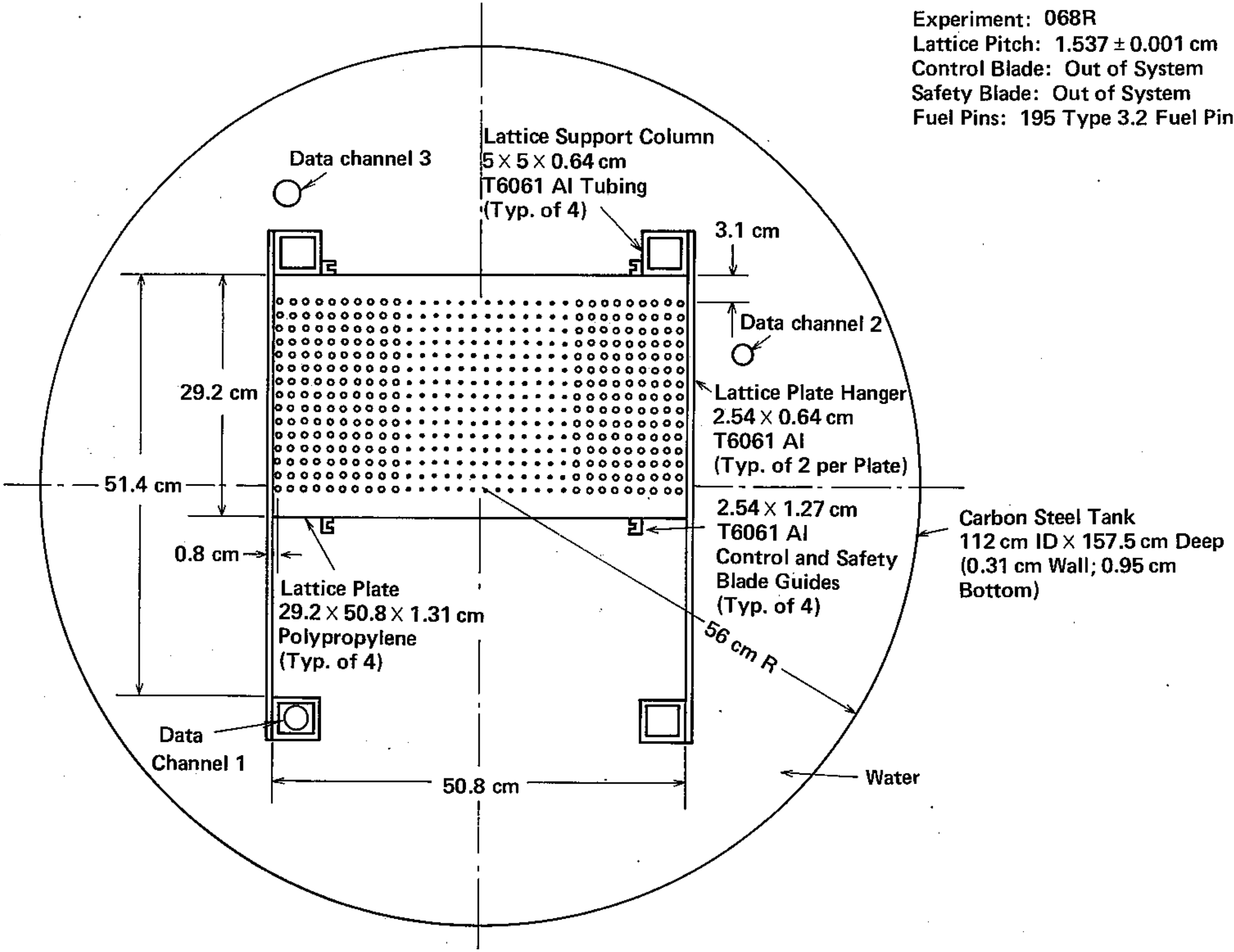
[*1] Calculation - Experiment

Table 7. K_{eff} of Water and Organic Moderated FTR Fuel Pins

Lattice Pitch (cm)	Vol. Ratio (V_m/V_f)	K_{eff} Water	(%ΔK)	Organic	(%ΔK)
0.761	1.62	0.99476	(0.52)	0.99393	(0.61)
0.968	3.49	1.00089	(0.09)	1.00019	(0.02)
1.242	6.65	0.99861	(0.14)	0.99598	(0.40)
1.537	10.93	0.99770	(0.23)	0.99237	(0.76)
1.935	18.13	0.99910	(0.09)	0.99286	(0.71)

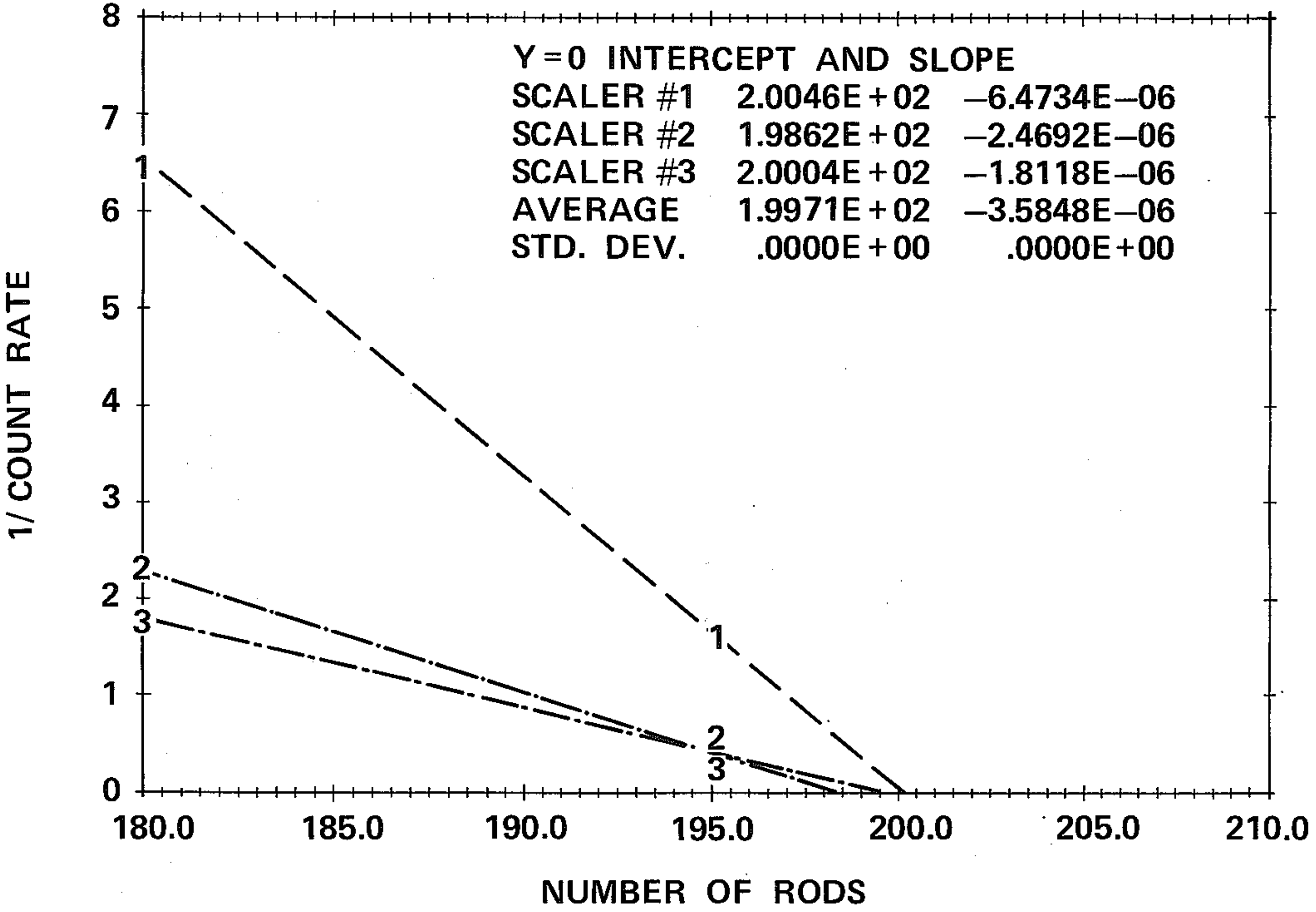
Table 8. Estimation of Different Chemical Bonding Model between Nelkin and Free Gas

Moderator	Lattice Pitch(cm)	K_{eff}			
		Nelkin	(%ΔK)	Free Gas	(%ΔK)
Water	0.968	1.00090	(0.09)	1.01364	(1.36)
	1.537	0.99770	(0.23)	1.01459	(1.46)
Organic	0.968	1.00019	(0.02)	1.01248	(1.25)
	1.537	0.99237	(0.76)	1.00936	(0.94)



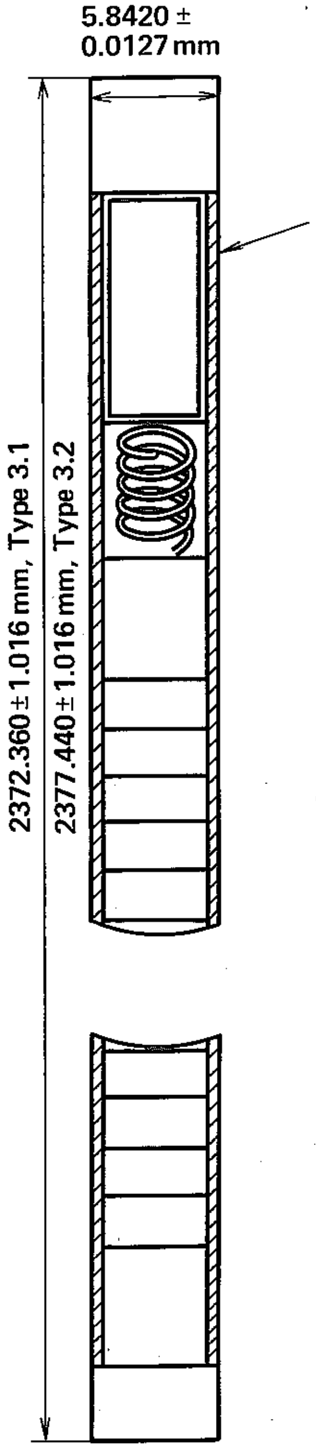
Experiment: 068R
Lattice Pitch: 1.537 ± 0.001 cm
Control Blade: Out of System
Safety Blade: Out of System
Fuel Pins: 195 Type 3.2 Fuel Pins

Fig. 1. Layout Showing Final Subcritical Loading Arrangement of Experiment 068R FTR Fuel Pins Immersed in Water Moderator



CFRP−PNC−FTR−068R July 7, 1985−1/Count Rate Approach

Fig. 2. CFRP−PNC−FTR−068R JULY 7. 1985 TYPE 3.2 FTR FUEL PINS. 20.0C FULL WATER REFLECTION 1.573CM SQUARE PITCH LATTICE 15 FUEL PINS WIDE



	TYPE 3.1	TYPE 3.2
Wt% ^{238}Pu	0.0130	0.0111
Wt% ^{239}Pu	21.0641	17.1251
Wt% ^{240}Pu	2.8606	2.3150
Wt% ^{241}Pu	0.3461	0.2876
Wt% ^{242}Pu	0.0526	0.0381
Wt% ^{241}Am	0.0504	0.0597
Wt% ^{235}U	0.4547	0.4853
Wt% ^{238}U	63.5857	67.8638
O	11.5728	11.8143
g/cm^3 Pellet	9.903	9.994
g/cm^3 Stack	9.783±0.318	9.830±0.320

Weighted average Am-241 content as of January 1, 1976, based on measured data from various pellet lots.

Fig. 3. Simplified Description of FTR Fuel Pin

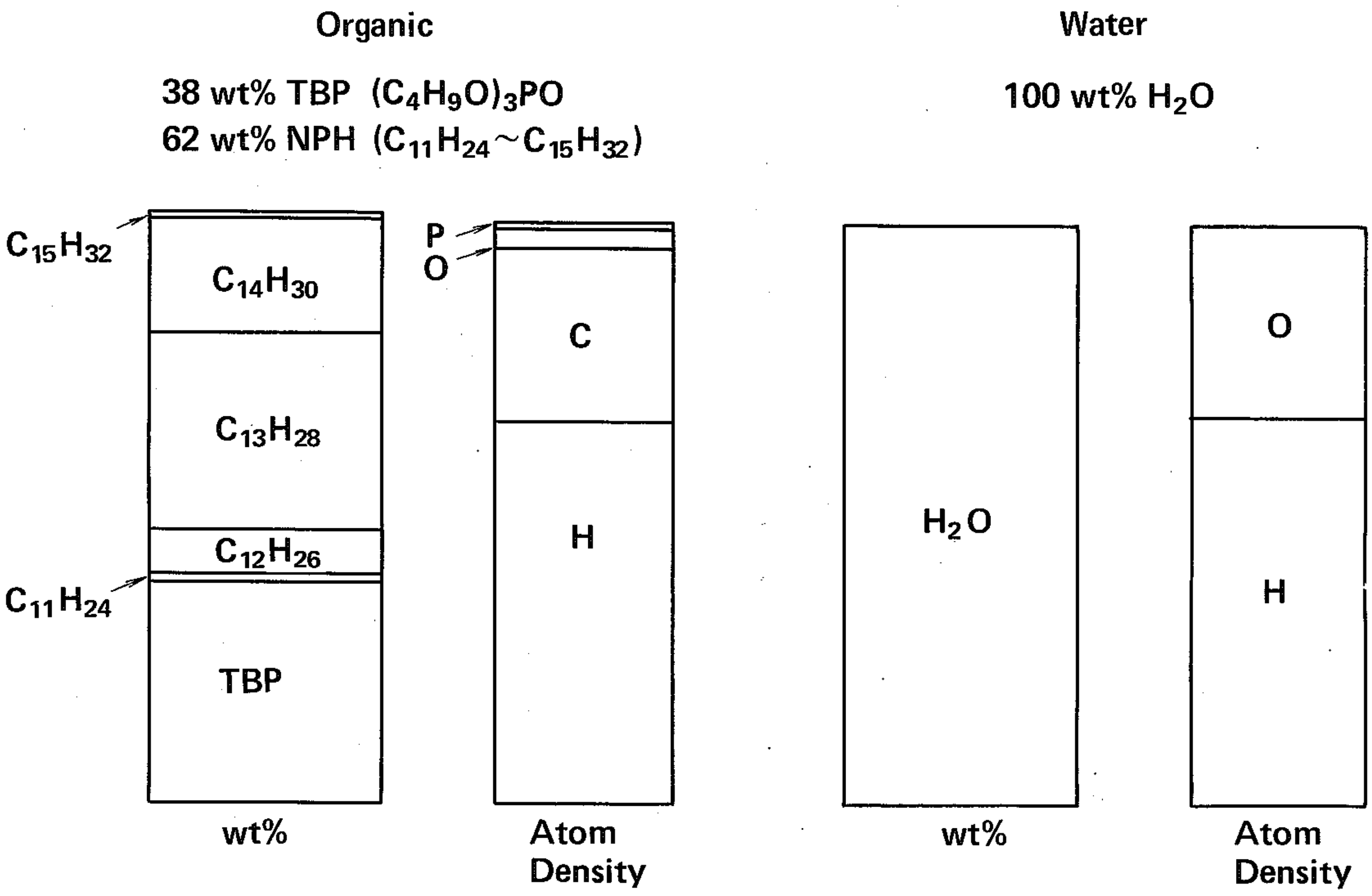


Fig. 4. Comparison of Organic and Water Composition

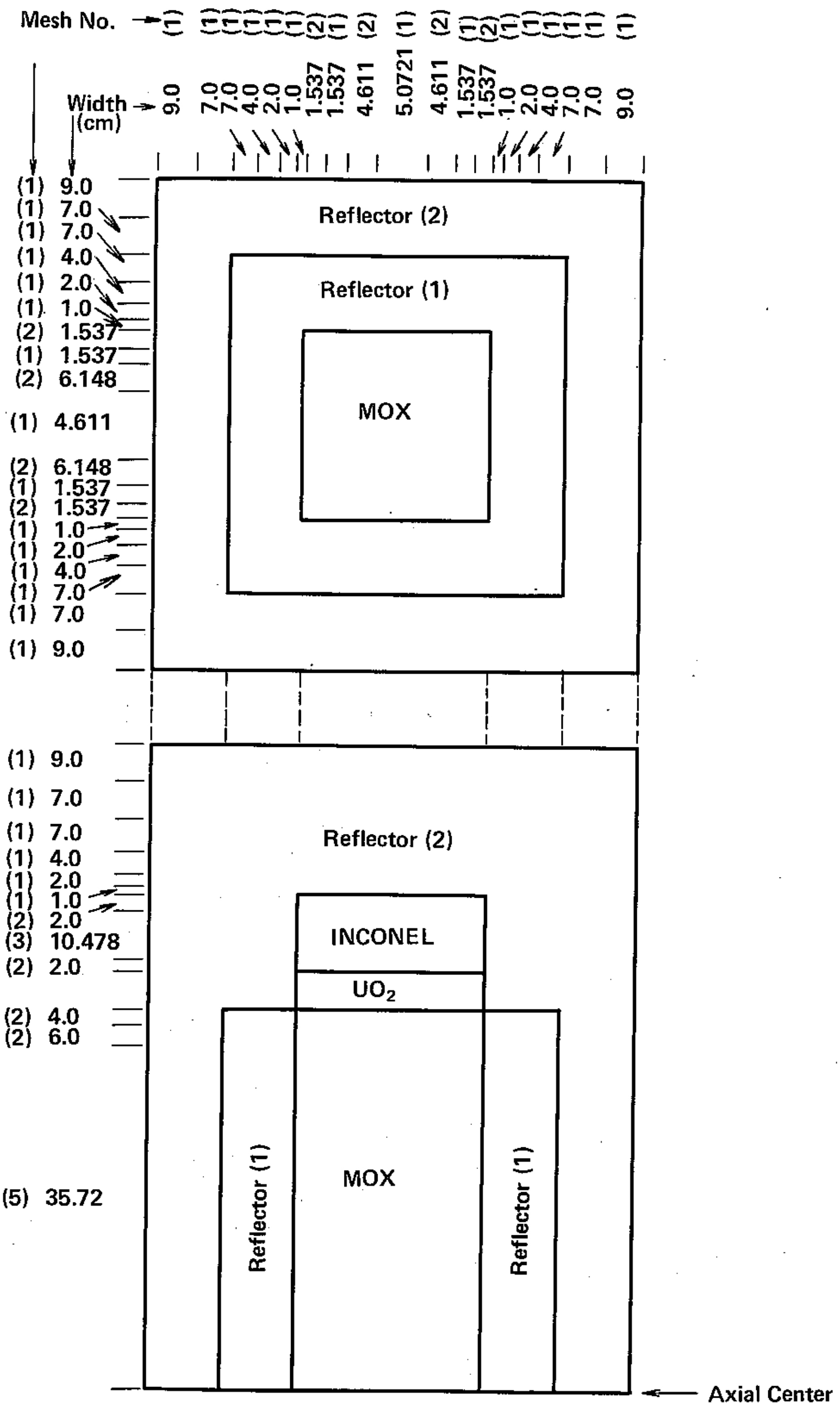
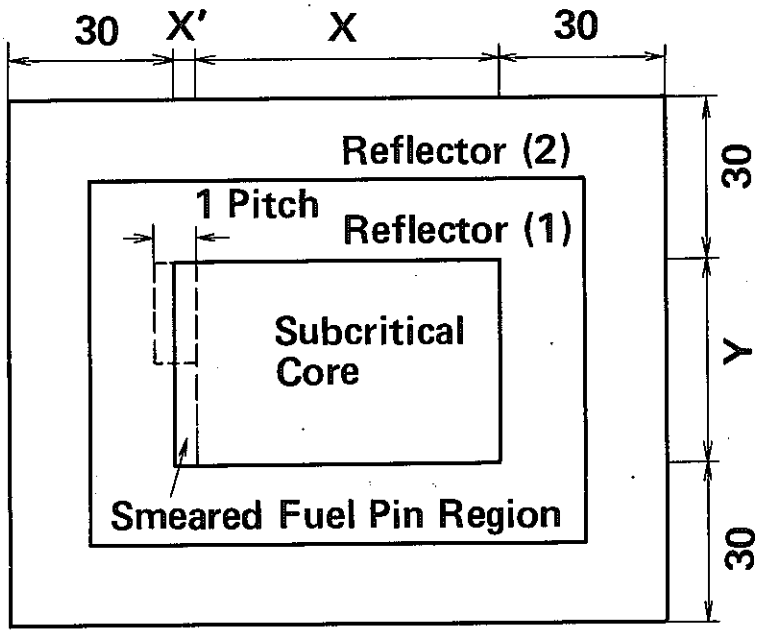


Fig. 5. Geometrical Model of X–Y and Axial Direction (Lattice Pitch : 1.537 cm)

Experimental Critical Fuel Pins – Loaded Subcritical Fuel Pins

⟹ Smeared Fuel Pin to the Subcritical Core



Critical Core Configuration

(Unit : cm)

Fig. 6. Critical Core Configuration

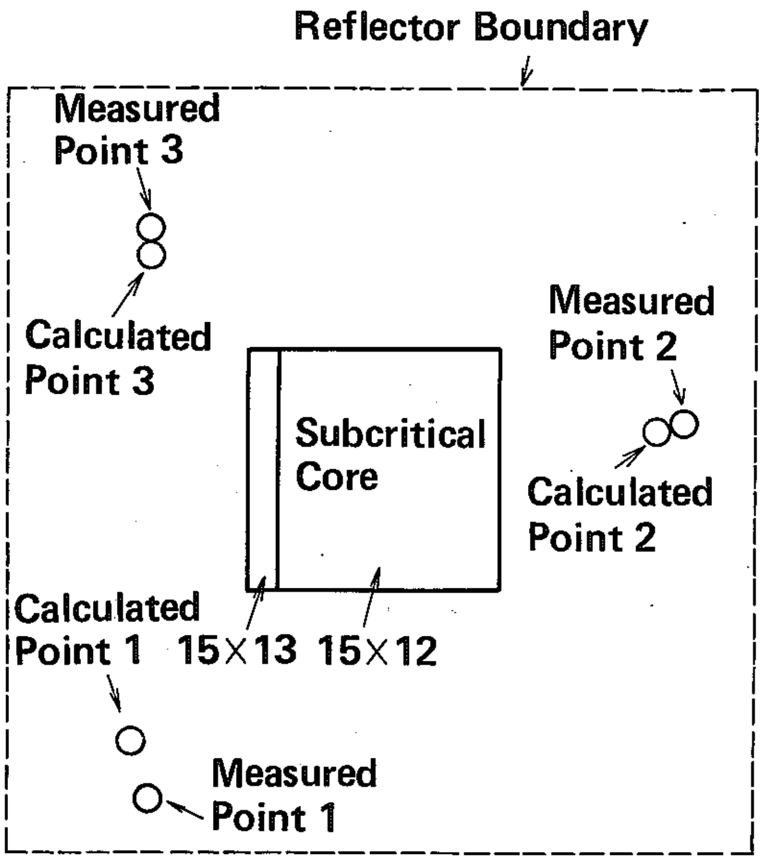


Fig. 7. Simulated Model of Inverse Neutron Count Measurement (Lattice Pitch : 1.537 cm)

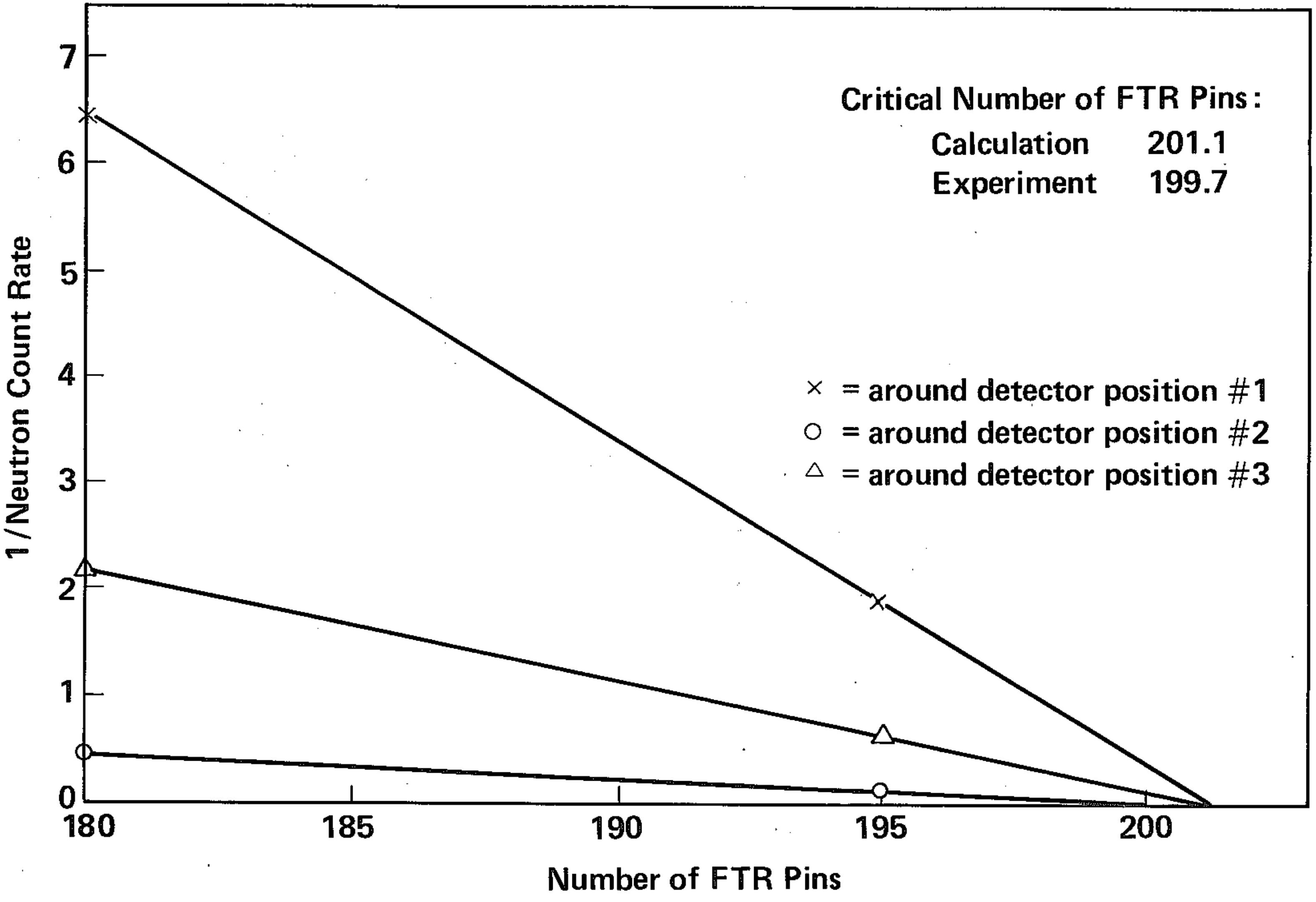


Fig. 8. Determination of Critical Number of Water Moderated FTR Pins with 1.537 cm Lattice Pitch

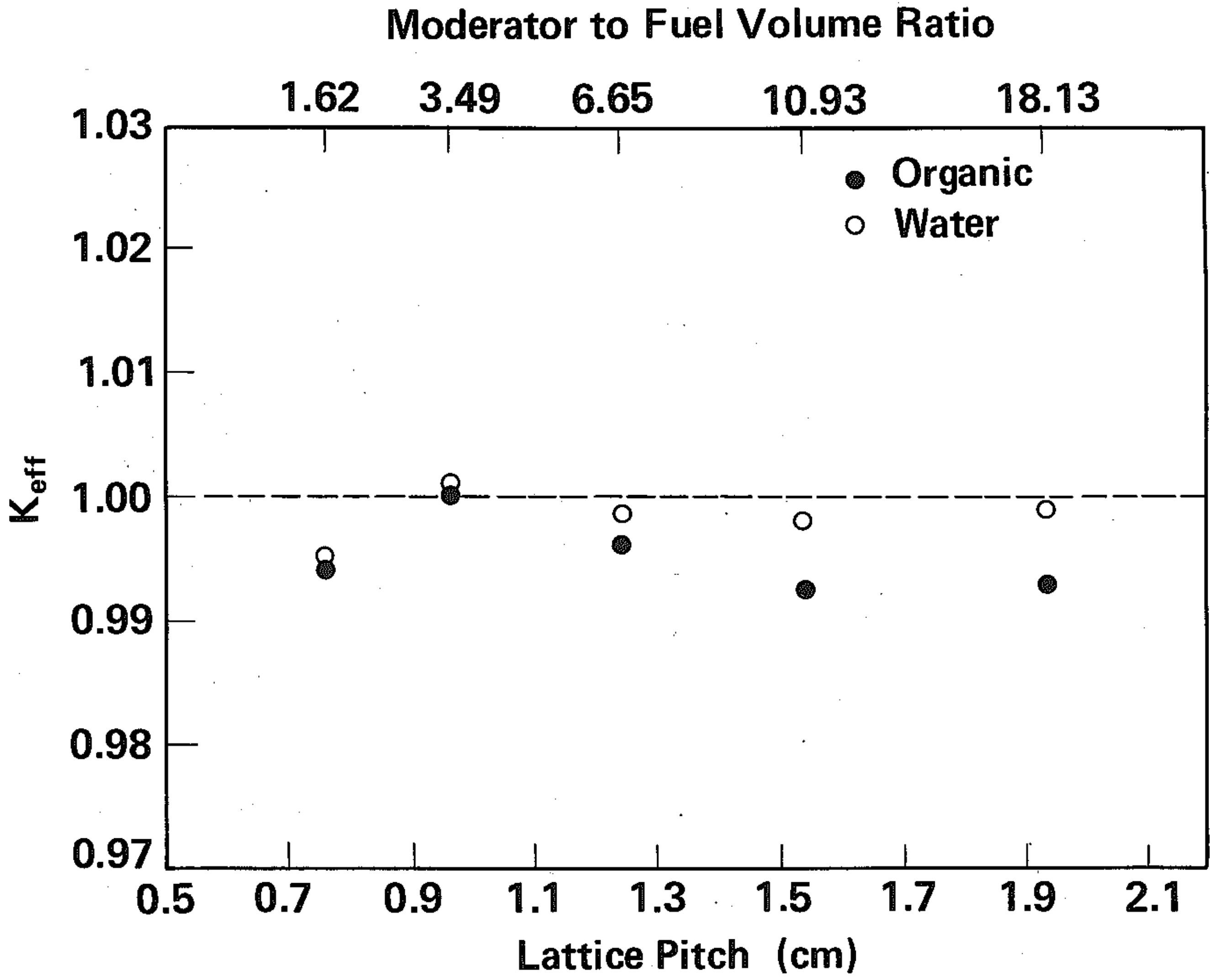


Fig. 9. Comparison of Benchmark Calculations between Organic and Water Moderated FTR Fuel Pins

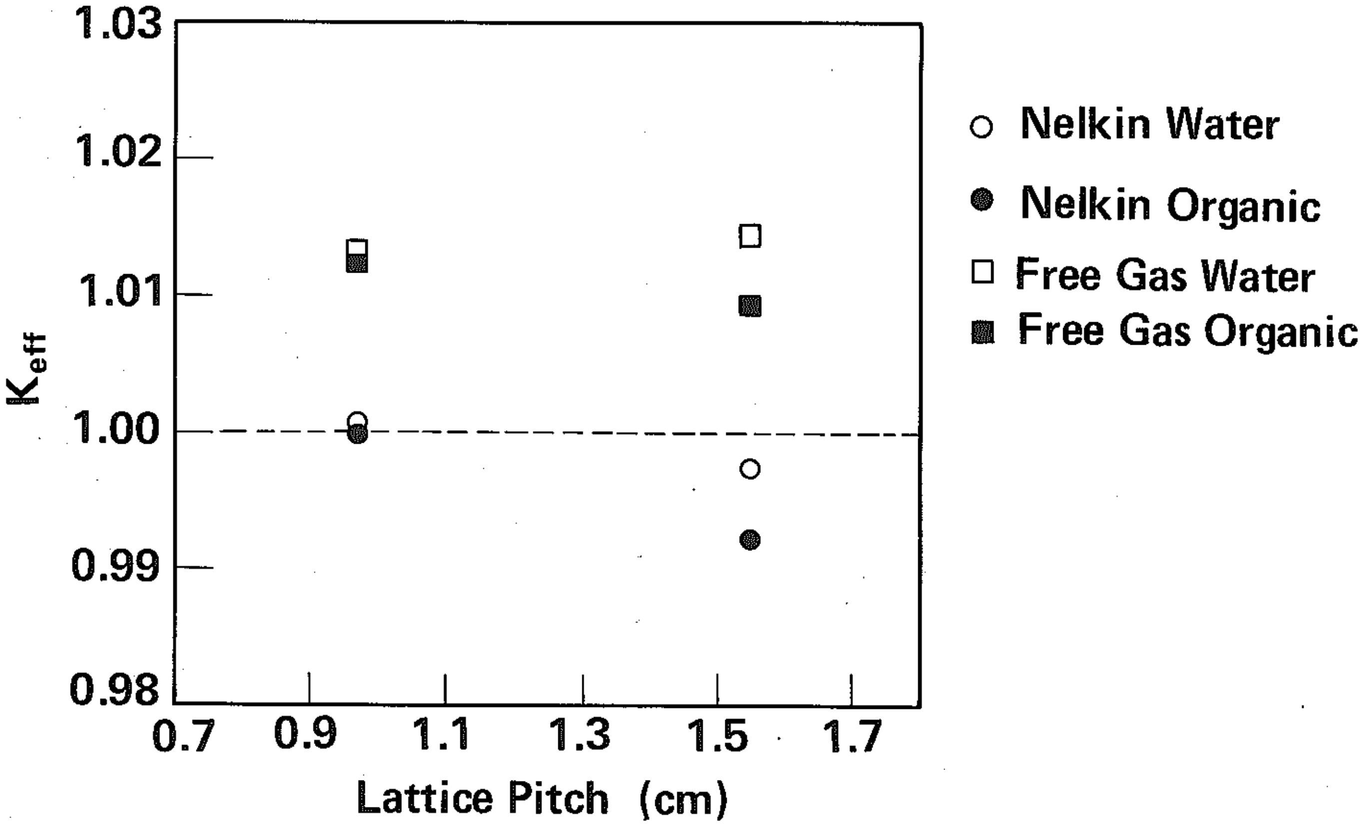


Fig. 10. Estimation of Different Chemical Bonding Model between Nelkin and Free Gas