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Presented by D. K. Butler

Measurements and Analysis of Al-, Al₂O₃-, and BeO-reflected Fast Critical Experiments

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1. Introduction

The Argonne nuclear rocket program was developed as a backup effort to the graphite rocket reactor program at Los Alamos. As an alternate to the graphite system, Argonne recommended a refractory-metal-based fast reactor, since the refractory metals have potential for operation at very high temperatures. A fast-neutron spectrum would make it possible to use materials with high thermal-neutron cross sections, and to obtain cores of any desired size down to 20 liters or less. Initial studies led to the choice of tungsten as the refractory metal, possibly alloyed with rhenium where added strength or temperature performance was required, and of uranium oxide as the nuclear fuel. The studies also showed that the best system would probably have a reflector that would help flatten the power distribution and improve neutron economy. With weight at a premium the use of light reflector materials, including aluminum oxide, beryllium, and beryllium oxide, was considered most promising. The series of critical assemblies discussed here was performed in connection with the program for development of a fast tungsten reactor for nuclear propulsion.

At the time, almost nothing was known about the neutronics of such systems. A program of critical experiments was therefore planned to obtain some basic information about fast tungsten-based systems. The approach selected was to begin with systems which had already been studied, such as Assemblies 11 and 22 of ZPR-3 and Assembly 1 of ZPR-6. A series of critical configurations was constructed in which only a single change of composition was made from one assembly to the next.

The experiments were performed with the split-bed assembly ZPR-9. The cores were made as close approximations to cylinders as possible in the rectangular drawers of the assembly. Criticality was normally achieved by varying the core radius.

In each assembly a number of measurements were made. In addition to criticality, determinations of fission-rate distribution, neutron lifetime, and central

and spatial reactivity worths were made. Some time was also spent studying properties related to control, including the effect of inserting a ring of boron into the reflector near the core boundary.

Many of the details of the experiments and some of the calculations have been reported previously.⁽¹⁻⁵⁾ Final reports summarizing all experimental results are in preparation.

Rather than go over all details of measurements, we have tried to select some results for which we have made theoretical comparisons and which, we think, show what has been learned and what is yet to be learned.

Included for discussion will be: (1) criticality, (2) the spatial distribution of fission rates, and (3) the worths of various materials at the core center and as a function of radius.

2. Description of Assemblies

The critical configurations determined in the experiments are shown in Tables 1 and 2. The dimensions have been adjusted to smooth the actual "stair step" outline of the experimental radial outline.

The columns in Table 2 labeled core material and reflector show the progression of materials changes. Assembly 1 is the assembly that had the same core that was studied in ZPR-6 and ZPR-3, but instead of the dense U²³⁸ reflector of the previous experiments, dense aluminum metal was substituted. Assemblies 2, 3, and 4 represent stepwise replacement of U²³⁸ by tungsten. In Assembly 4 the replacement was complete.

Tungsten is a strong resonance absorber of neutrons. Assembly 5 was planned to have a somewhat softer spectrum produced by the addition of some carbon to the core. The actual amount of carbon added was only enough to produce a moderate spectral softening. The sixth assembly was made more like an actual rocket system, by simulating void with perforated aluminum plates. The void would correspond to propellant channels in the actual system.

The first six assemblies had dense aluminum metal reflectors. For the seventh assembly, the reflector was

TABLE 1. PHYSICAL DESCRIPTION OF ZPR-9 ROCKET ASSEMBLIES

	Assembly No.								
	1	2	3	4	5	6	7	8	9
Core									
Radius (cm)	31.39	33.74	35.50	24.62	33.89	36.60	27.56	31.71	26.13
Height (cm)	51.10	61.26	71.42	56.18	56.18	61.26	51.10	51.10	51.10
Inner Radial Reflector									
Radius (cm)	66.63	70.28	70.56	65.37	70.83	75.64	65.52	42.54	77.18
Height (cm)	51.10	61.26	71.42	56.18	56.18	61.26	51.10	51.10	127.30
Outer Radial Reflector									
Radius (cm)	—	—	—	—	—	—	77.18	88.95	88.95
Height (cm)	—	—	—	—	—	—	112.06	51.10	127.30
Axial Reflector									
Thickness (cm)	30.0	30.0	30.0	30.0	30.0	38.10	30.48	37.95	38.10
Core Composition (atoms/cm ³ × 10 ²⁴)									
U-235	0.004516	0.004495	0.004473	0.006717	0.004486	0.004519	0.004498	0.004508	0.004497
U-238	0.03561	0.02418	0.01708	0.000485	0.000323	0.000346	0.000325	0.0003259	0.0003255
C/O	—	—	—	— Carbon	0.02972	—	— Oxygen	0.008863	0.008984
Al	0.006784	0.006803	0.006501	0.0067	0.006803	0.01549	0.01594	0.01656	0.0177
W	—	0.0122	0.02129	0.03953	0.02134	0.02152	0.02169	0.02138	0.02126
Fe	0.000258	0.000221	0.000202	0.000415	0.00221	0.000225	0.000298	0.0002556	0.0003337
Inner Radial Reflector (atoms/cm ³ × 10 ²⁴)									
Be	—	—	—	—	—	—	—	0.05809	—
O	—	—	—	—	—	—	0.05449	0.05809	0.05477
Al	0.05658	0.05658	0.05658	0.05658	0.05658	0.05658	0.04324	0.006917	0.04139
Outer Radial Reflector (atoms/cm ³ × 10 ²⁴)									
Al	—	—	—	—	—	—	0.05658	0.0572	0.0573
Axial Reflector (atoms/ cm ³ × 10 ²⁴)									
O	—	—	—	—	—	—	0.05449	—	0.05477
Al	0.05658	0.05658	0.05658	0.05658	0.05658	0.05658	0.04324	0.0572	0.04139

changed to aluminum oxide. With the added scattering by the oxygen, one can see by the change in critical core volume that this was a much better reflecting material. The original plan was to have only one more assembly (No. 8), namely, one with a beryllium oxide reflector. The delay in obtaining safety approval combined with available time made it possible to insert Assembly 9, which had oxygen added to the core to simulate uranium oxide. This was actually done in the critical experiment by replacing some of the perforated aluminum plates with aluminum oxide. The final assembly of the list had a beryllium oxide radial reflector. In order to simplify obtaining safety approval for operation, aluminum metal was used as the axial reflector to insure a negative reactivity coefficient for axial expansion.

3. Criticality

Table 2 shows results of calculations of k_{eff} for the various assemblies, made by conventional techniques of neutron diffusion and transport theory. For convenience the cross-section set of Hansen and Roach⁽⁷⁾

TABLE 2. VALUES OF k_{eff} CALCULATED BY VARIOUS METHODS

Assembly	Core Materials	Reflector	Core Volume, liters	k_{eff} Diffusion	k_{eff} S_4	k_{eff} 26 Groups
1	U ²³⁵ , U ²³⁸	Al	158	0.966	0.980	0.989
2	U ²³⁵ , U ²³⁸ , W	Al	219	0.975	0.987	0.992
3	U ²³⁵ , U ²³⁸ , W	Al	283	0.976	0.987	0.988
4 ^(b)	U ²³⁵ , W	Al	107	0.982	1.004	0.996
5	U ²³⁵ , W, C	Al	202	0.998	1.020	1.007
6	U ²³⁵ , W ^(a)	Al	258	0.999	1.024	1.018
7	U ²³⁵ , W ^(a)	Al ₂ O ₃	117	0.995	1.003	0.984
9	U ²³⁵ O ₂ , W ^(a)	Al ₂ O ₃	110	1.004	1.012	(0.992)
8	U ²³⁵ O ₂ , W ^(a)	BeO-Al	161	1.007	1.021	0.991

^(a) Core diluted to simulate porosity.

^(b) Fuel concentration 50% greater than for other assemblies shown.

were used early in the program. These did not contain tungsten so that a set of tungsten cross sections were assembled from whatever microscopic information was available.

Several cross-section sets were generated from some basic data. On the basis of the first four critical as-

semblies, one cross-section set was selected as giving the best agreement with experiment. In using this cross-section set to calculate properties of the later assemblies, we have continued to get good results.

Cross sections for rhenium were assembled in a similar way. When used in calculations, these gave results in poorer agreement with the measurements.

Throughout the early part of the rocket critical program, we attempted to calculate properties of tungsten-based systems by one-dimensional diffusion and transport theory. One approach was to calculate in spherical geometry and use a shape factor to convert to cylindrical geometry. For shape factors we were forced to rely on data obtained with uranium reflectors. A second approach was to use the usual cylinder-and-slab synthesis in which the buckling obtained from a slab problem is used in the succeeding cylindrical calculation (or vice versa). It was clear that the light reflectors returned low-energy neutrons distributed spatially in a way that violated the separability of variables assumed in this calculational technique.

Only recently has a many-group two-dimensional diffusion-theory code become available. The column labeled k_{eff} diffusion in Table 2 shows the k_{eff} obtained by use of this code. These values proved to be far more consistent with experiment than those from the one-dimensional iteration procedure.

The calculations for the nine assemblies shown have a rms deviation from 1 of 1.8%. The greatest discrepancy can be seen to be associated with the system containing U^{238} . The U^{238} cross-section set that was used results in underprediction of k . Taking the last six assemblies separately, the rms deviation is only about half that of the U^{238} diluted assemblies.

Even though they were of solid metal, the aluminum reflectors of the earlier assemblies were not good reflectors of neutrons. This means that it is likely that they will not satisfy the diffusion approximation very well. In order to check this, a series of S_N transport calculations were made. These were made in one-dimensional spherical geometry, and the results were converted to equivalent change of k_{eff} as shown in the column of Table 2 labeled k_{eff}, S_4 . As a group, these values show a rms deviation from 1 to 1.6% in Δk . As might be expected, the results for the first six aluminum-reflected assemblies were from 1 to 2% more reactive than would be indicated by results of diffusion theory. The corrections by transport theory for Assemblies 7, 8, and 9 were less because of the greater scattering power of the oxides.

By 1965 there were available considerable new basic data on tungsten and other materials of interest in rocket systems. In order to see whether these new data could be used to obtain improved calculational re-

sults, a 26-group cross-section set was assembled, using the MC² code.⁽⁶⁾ Unfortunately, the library available did not have complete cross sections for very low energies, so that it proved necessary to continue use of the older Hansen-Roach cross sections below 30 eV. Values for k_{eff} calculated by two-dimensional diffusion theory are shown in the last column. As a group, these cross sections show the best accuracy; however, this improvement is primarily in the calculation of the first three assemblies for which the new U^{238} cross-section set gives improved results. Results for tungsten-based systems with the new set are actually a little poorer on the average than are the results based on the Hansen-Roach set.

It is perhaps appropriate to point out that these assemblies are no different from other fast assemblies in that the uncertainty in critical volume is typically something like five times as great as the uncertainty in k . To a person loading a critical assembly and working with fuel of essentially fixed enrichment, this can result in a considerable problem. On the other hand, the uncertainty in enrichment is only about twice that in k . Therefore, an uncertainty in critical mass figured on the basis of uncertainty of enrichment rather than volume as the desirable feature is less sensitive to the geometry of the system and hence allows for a better comparison of the ability to calculate different sized systems.

4. Fission Rates

The radial distribution of fission rates at the core midplane was studied, both theoretically and experimentally. Figure 1 shows the results for the U^{235} fission rate in Assembly 7, the assembly with an aluminum oxide reflector. The values are normalized to unity at the core center. Three sets of data are plotted. The triangular dots were obtained from foil irradiations. The squares were measurements with a solid-state fission counter, and the small crosses show the results of a calculation by one-dimensional cylindrical diffusion theory.

All measurements agree rather well except in the reflector beyond 60 cm, where the measured values are higher than the calculated values. This may be primarily due to the return of low-energy neutrons from the materials surrounding the assembly.

There is another significant disagreement at 30 cm, which amounts to about 30%. Part, if not all, of this discrepancy results from the fact that the counter traverse was made vertically through the core and the foil irradiation horizontally. The pieces of material from which the core is assembled are loaded vertically so there may be streaming up and down that does not exist horizontally.

FIG. 1. Radial Distribution of U^{235} Fission Rate at Axial Midplane of Assembly 7. FIG. 2. Radial Distribution of U^{238} Fission Rate at Axial Midplane of Assembly 7.

Figure 2 shows the U^{238} fission rate normalized to one at the core center for the aluminum oxide-reflected Assembly 7. The squares again represent the results obtained with a solid-state counter; the triangle are

results obtained with foils. Calculated results are shown as small crosses. In the reflector the fission rate depends on high-energy neutrons, and the discrepancy is quite acute. Out to about 45 cm, the major discrep-

TABLE 3. CENTRAL WORTHS (lh/kg)

Assembly	U ²³⁵	W	H
1	240 (276) ^(a)	-22.6 (-26.0)	
4	270 (286)	-19.6 (-22.3)	3300 (3500)
4B	219	-17.0	-2900
5	197 (221)	16.3 (-21.0)	3900 (600)
6	148 (140)	-10.6 (-11.0)	3700 (2500)
7	176 (163)	-11.0 (-12.8)	3300 (2900)
9	207 (195)	-15.9 (-16.3)	4900 (2100)
8	169 (169)	-11.4 (-14.0)	4800 (1800)

^(a) Calculated values in parentheses.

ancy is between the foil irradiations and the solid-state counter results, with the latter following the calculations quite closely. Beyond 45 cm, both sets of experimental results are much higher than the calculated values. It is not likely that this was caused by room return, since not many high-energy neutrons are returned from the room. As with U²³⁵, some of the discrepancy may be due to streaming in the vertical direction. But it is the foils, which should be less effected by streaming, that have the higher relative count rate. Transport theory might give better agreement with experiment. Clearly, this is the situation leaving something to be learned about both experimental and calculational techniques.

5. Reactivity Traverses

The third series of experiments and calculations are those related to the determination of the reactivity worth of various materials in the assemblies. In all of the assemblies, the worths of a wide variety of materials were measured at the core center. In Table 3 are summarized some of the central worth measurements. The figures in parentheses are the calculated values obtained with perturbation theory using the Hansen-Roach cross-section set. The agreement between experiment and theory for U²³⁵ is reasonably good but some discrepancy remains. The results for tungsten show good agreement between theory and experiment, although again the agreement is not as good as to achieve a thoroughly reliable calculational technique.

The measurements with hydrogen were made to determine the effects of hydrogen propellant on the core. In the fast cores listed in this table the introduction of hydrogen increases reactivity in all but one instance, Assembly 4B. This was a variation of Assembly 4 in which less than 4 v/o rhenium was added to the core. The addition of rhenium increased the critical volume about 30% over that of Assembly 4. This affected hydrogen worth by reversing the sign though maintaining about the same magnitude of reactivity effect. In rocket design this could be a very significant consideration. It shows how reactivity effects can be appreciably sensitive to small changes in core composition.

The calculated worths of hydrogen shown in the third column are in serious disagreement with the measured values. The indications are that the U²³⁵ cross sections tend to overpredict reactivity, with the tungsten producing a compensating underprediction.

In the later assemblies a series of spatial reactivity worth distributions were measured. The experimental technique used was simple in principle. A small tube was installed through the center of the assembly along a diameter at the midplane. Small samples of material were removed through this tube and the resulting changes in reactivity recorded. The key feature in this experiment was the use of a sample drive system that transmitted signals indicating sample position to a computer and the simultaneous transmission to the computer of the instantaneous neutron level. The computer was programmed to calculate the instantaneous reactivity and store it as a function of sample position. The results from several sample traverses could be directly combined. The computer would then directly give output reactivity versus sample position.

The samples used were small and thus the reactivity changes were small. Repeated measurements at neutron levels somewhat higher than for normal operation were required to give results relatively free of reactor noise. One uncertainty in any traverse experiment of this type is the effect of neutron streaming through the traverse hole. For these experiments the hole was about 1 cm in diameter, and some effects of streaming could be expected over distances of perhaps 1 or 2 cm for fast neutrons. With aluminum or beryllium oxide reflectors more serious streaming problems might arise because of the thermal neutrons in the reflector region.

An experimental check of streaming was made by filling the hole both in front of and behind the sample with a lead rod.

Figure 3 shows the results obtained with a sample of U²³⁵. The heavy solid curve was obtained with samples in the empty traverse tube. The lighter line was

FIG. 3. Radial Distribution of Worth of U^{235} at Core Midplane in Assembly 8. Thin line represents results obtained with lead rod.

FIG. 4. Radial Distribution of Worth of Tungsten at Core Midplane in Assembly 8. Solid curve shows experimental data, points results of calculation.

FIG. 5. Radial Worth Distribution of Rhenium at Core Midplane of Assembly 8.

FIG. 6. Radial Worth Distribution of B¹⁰ at Core Midplane in Assembly 8.

obtained with the lead scattering rod preceding and following the sample. The agreement is very close. Another measurement with B^{10} showed similar good agreement. The small irregularities in the traces result from reactor noise.

Figure 4 shows results for a traverse of a sample of tungsten. These measurements were made in Assembly 8 after a small amount of hydrogen was added to the core and reflector in order to study the space-dependent reactivity change with the addition of hydrogen. The hydrogen made a very small change in the other properties of the system. The measured reactivity worth is everywhere negative. A fairly sharp minimum in the beryllium oxide reflector results from the significant number of low-energy and thermal neutrons. Also shown are values calculated using diffusion perturbation theory. Calculations shown as the crosses were made with the Hansen-Roach set, labeled 201, and the squares represent calculations made with the 26-group set, labeled 830. Both cross-section sets give values which are too negative, though the 26-group set falls a little closer to the measured value. At least part of the discrepancy is probably due to the self-shielding effect of the tungsten sample (in the form of a solid cylinder of about 50 g). One problem with these measurements is determining the magnitude of sample self-shielding.

Figure 5 shows results obtained by traverses of rhenium samples in the same assembly. Two experimental curves are shown by the continuous lines. The one with the smaller negative specific worth was obtained with a 54.9-g sample; that with the greater worth was obtained with a 3.4-g sample. Again calculated values are shown. In this instance the rhenium cross section assembled from the Hansen-Roach set leads to an underestimation of the effect while the 26-group set very much overestimates it.

Figure 6 shows results for B^{10} . Four experimental measurements are shown, again representing different sample sizes. The uppermost curve was obtained with a sample of 0.775 g, the second with one of 0.113 g, the third with one of 0.045 g, and the bottom curve for a sample of 0.009 g. Again the results of calculations with both cross-section sets are very much too negative in the beryllium oxide reflector. In this instance the series of samples indicates that the value for an infinitesimal sample, which would satisfy the perturbation approximation, probably would not be nearly as negative as the calculations.

6. Conclusions

As mentioned in Sect. 1, many additional results for these and other qualities were measured and calculated. Although they indicate that the neutronic prop-

erties of the major constituents in the core center are reasonably well predicted, the properties of hydrogen and rhenium are less well predicted. Away from the center of the core the predictions are less good, especially for the Al_2O_3 - and BeO -reflected systems. The disagreements are consistent with the uncertainty of the cross-section information used. Improved cross sections could be expected to give better agreement with experiment. It is possible that this would also require local flux averaging of the cross sections in the various regions where the spectra differs so greatly.

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Discussion

Mr. Richards (ANL): Have you thought of replacing tungsten by molybdenum? Molybdenum is very similar to tungsten, somewhat lighter, and possibly easier to manufacture.

Mr. Butler: In the early parts of the program there was a study of molybdenum, tungsten, and other materials. We have not made measurements with molybdenum. As I remember, molybdenum does not quite have the temperature potential, so we turned directly to tungsten. I did make one calculation, again using the Hansen-Roach molybdenum data, and we decided that if we needed additional reactivity in these cores we might use molybdenum at the cold ends. Molybdenum is neutronicallly a better material.

Mr. Richards: The temperature properties of molybdenum are very similar to tungsten except for extremely high temperatures.

Mr. Butler: For rocket applications the interest is in the highest temperature attainable.

Mr. Meister (Karlsruhe): Did you compare the

ratio of reactivity worths of boron to U^{235} with the Hansen-Roach calculations, or make some lifetime measurements?

Mr. Butler: Yes we have made such calculations, and we also have some lifetime measurements. These will be included in the Proceedings.

Mr. Becker (RPI): Would you describe the experimental procedure used to measure the reactivity?

Mr. Butler: The technique was that developed by C. E. Cohn of Argonne. The sample location and flux as a function of time are read into a DDP-24 computer, programmed to calculate the instantaneous re-

activity on the basis of what the reactivity was a short time before. It continuously calculates reactivity changes and then interprets these as absolute reactivity and stores them as such.

Mr. Becker: What I am curious about is whether or not there was any difference in the core lifetime as you move the perturbation from the core into the reflector. For example, if it depressed the flux in the reflector it might affect the core lifetime. Was that considered in the reactivity calculation?

Mr. Butler: I suggest that you contact Mr. Cohn for such details.