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**HYDROGEN MODERATION -
A PRIMARY NUCLEAR SAFETY CONTROL
FOR HANDLING AND
TRANSPORTING LOW-ENRICHMENT UF₆**

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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**UNION
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**UNION CARBIDE CORPORATION
NUCLEAR DIVISION**

Operating the

- OAK RIDGE GASEOUS DIFFUSION PLANT
- OAK RIDGE Y-12 PLANT
- OAK RIDGE NATIONAL LABORATORY
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UNION CARBIDE CORPORATION

NUCLEAR DIVISION

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Abstract

The use of hydrogen moderation as a primary nuclear safety control has been in effect for many years for large-scale UF_6 shipments among AEC-owned contractor operated facilities. Since it appears that the extension of the use of moderation controlled shipments to licensee as well as contractor organizations is desirable, this report summarizes, in support of the method, the work done in developing basic criticality data and the necessary equipment and operational techniques.

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Introduction

It may be stated a priori that the continued growth of the nuclear industry is closely allied with its ability to reduce the costs of fuel fabrication and reprocessing for reactors, including handling and transportation charges of the basic fuel, uranium hexafluoride. One of the principal methods now available for reducing these latter charges is through the bulk shipment of low-enrichment UF_6 , using limited moderation as the primary nuclear safety control.

Moderation control has been long recognized as an acceptable method of establishing nuclear safety in the transportation of radioactive materials,¹ having been used for many years in the large-scale shipment of UF_6 among AEC-owned contractor operated facilities. Since it appears that the extension of the use of moderation controlled shipments to licensee as well as contractor organizations is desirable, this report summarizes, in support of the method, the work done in developing basic criticality data and the equipment and operational techniques that provide necessary controls.

HYDROGEN MODERATION - A PRIMARY NUCLEAR SAFETY CONTROL FOR HANDLING AND TRANSPORTING LOW-ENRICHMENT UF₆

GENERAL NUCLEAR SAFETY CONSIDERATIONS

Of all the factors affecting the neutron economy in a chain-reacting uranium system, the hydrogen moderation is perhaps the most important other than the actual enrichment of the fissile ²³⁵U isotope. Moderation may be defined as the slowing down of neutrons by collision processes from the high velocities and correspondingly high energies at which they are produced by fission, to the velocities and corresponding energies at which the probability of capture in a fissile atom is large. It is generally expressed as a ratio of the number of moderator atoms to fissile atoms in the fuel material, i.e., the H/²³⁵U atomic ratio.

Although all materials moderate neutrons to some degree, only those elements whose atomic weight is close to that of the neutron are considered to be good moderators, since the neutron energy loss per collision with such nuclei is relatively large. The critical mass of hydrogen-moderated uranium systems is quite small compared with that of unmoderated uranium. It is well known that less than a kilogram of 93% ²³⁵U enriched uranium can be made critical in water solution, whereas over 100 kg of the corresponding unmoderated salt would be required. Similarly, the effect of hydrogen moderation on limiting critical enrichment values is also pronounced. In the case of UF₆, it appears that the maximum subcritical ²³⁵U enrichment is about 7% while that of the hydrogen-moderated UF₆ is 1.0%.

Since some neutron energy degradation occurs even in uranium metal systems, whose neutron chain is essentially dependent upon fast fission, it is apparent that in principle it would be impossible to have a truly unmoderated system of fissionable materials. In practice, however, an unmoderated system is generally referred to as one whose H/²³⁵U is zero. Because of the high efficiency of hydrogen as a neutron moderator and its overwhelming propensity in general industrial and community usage, chiefly in the form of water, nuclear safety criteria are usually based on hydrogen-moderated systems. However, it should be noted that hydrogen moderation is readily controllable by equipment and procedures that are not uncommon to normal industrial practice. This is particularly true for the UF₆ fuel whose chemical and physical properties permit the ready detection of hydrogenous contaminants.

NEUTRON PHYSICS OF HYDROGEN-MODERATED URANIUM SYSTEMS

As background for a discussion of moderation control, some of the more important criticality parameters will be reviewed briefly. First is the infinite medium multiplication factor, k_{∞} , since the primary requirement for criticality is that the k_{∞} of the fuel be greater than unity. In addition to delineating criticality limits, k_{∞} is a useful parameter in determining neutron interaction criteria.²

The Infinite Medium Multiplication Factor

The importance of hydrogen moderation is seen in a plot of the k_{∞} of the $\text{UO}_3\text{-H}_2\text{O}$ system, which is considered directly applicable to the $\text{UF}_6\text{-H}_2\text{O}$ system because of the similarity of the oxygen and fluorine cross sections (Fig. 1). It is noted from Fig. 1 that criticality would not be considered possible for homogeneous, hydrogen-moderated uranium materials if the ^{235}U enrichment were $\leq 1.0\%$ or at any enrichment if the H/U atomic ratio were ≥ 2200 .^{*} More pertinent however is the undermoderated region of the graph where the H/U ratio is close to zero. As indicated previously, it appears that criticality is not possible for unmoderated UF_6 at ^{235}U enrichments less than about 7%. This view was based upon an early computational survey³ which was supported by neutron multiplication measurements with large subcritical assemblies containing up to 20,000 lbs UF_6 whose H/ ^{235}U was 3.7 and ^{235}U enrichment 2.0%.⁴ In addition, exponential experiments⁵ with uranium metal of varying enrichments indicated the probable validity of the UF_6 determination.

The ^{235}U Critical Mass

Realizing, however, that the shipment of low-enrichment UF_6 in bulk quantities would become an important factor in the transportation of radioactive materials, it was deemed advisable to obtain integral critical mass data for UF_6 , not only to enable a more accurate assessment of moderation control procedures, but to provide additional data for nuclear safety guidance in diffusion plant operations.⁶ Summary results of these experiments,** performed at the Oak Ridge National Laboratory, are shown in Fig. 2.

^{*}The H/U ratio is the product of the H/ ^{235}U ratio and the ^{235}U enrichment.

^{**}Due to inherent difficulties of direct UF_6 criticality experiments, the tests were conducted with blocks of compressed UF_4 and CF_2 whose uranium density is comparable to the solid UF_6 , 3.2 g U/cc.

k_{∞} OF UO_3 (X) H_2O SYSTEMS FOR U(1) TO U(93)

Figure 1

The ^{235}U critical mass is plotted as a function of enrichment (Fig. 2) both for unmoderated UF_6 and for moderated UF_6 whose H/ ^{235}U ratio is 10. The latter value is considered to be a conservative upper limit of the hydrogen moderation found in UF_6 produced at gaseous diffusion plants. The enrichment values, indicated by dashed lines, are maximum subcritical enrichment limits. It may be of interest to note that some of these data represent perhaps the largest quantities of UF_6 ever made critical under controlled laboratory conditions; i.e., it was necessary to assemble 3490 kg U(18.8) F_6 in a water reflected, cubical configuration for criticality.

^{235}U CRITICAL MASS OF UF_6

Figure 2

Subcritical Enrichment Maxima

The effect of hydrogen moderation on the maximum subcritical enrichment of the $\text{UF}_6\text{-H}_2\text{O}$ system, $k_\infty = 1$, is shown by the curve in Fig. 3 which was derived from experimental and calculational results. The calculations were made using the ANISN transport theory code⁷ and the Hansen-Roach 16-group cross section set with a semiempirical modification of the ^{238}U absorption in the resonance region (Appendix I). The new value of $\text{U}(7.7)\text{F}_6$ for unmoderated uranium hexafluoride is slightly higher than the original determination of $\text{U}(7.0)\text{F}_6$. This difference may possibly be due to the lack of adequate inelastic scattering cross sections for fluorine in the early work.³ The minimum value of the curve in Fig. 3 (1.03% ^{235}U enrichment at an $\text{H}/^{235}\text{U}$ of 500) was determined with the Physical Constants Testing Reactor* at Hanford.^{8,9} The limiting $\text{H}/^{235}\text{U}$ of about 2200, corresponding to a concentration of about 12.1 g ^{235}U per liter of aqueous solution, was derived from experiments at ORNL.¹⁰

It may be noted that the shape of the curve clearly demonstrates the competing neutronic properties of hydrogen. As moderator is added to the UF_6 fuel, neutron kinetic energy is transferred to the hydrogen nucleus by elastic collision, slowing or moderating the neutrons to thermal energies, thus causing the curve to decrease initially. However, it is the radiative capture process $\text{H}^1(\text{n}, \gamma)\text{H}^2$ in competition with the ^{235}U fission process that causes the curve to increase at the higher moderation values.

Moderation Control Defined

The minimum value of the curve in Fig. 3, which occurs at 1.03% ^{235}U enrichment, could serve very well to differentiate between two important nuclear safety parameters, moderation control, and concentration control. The latter is sometimes regarded as a special case of the former since the ^{235}U isotope is the limited quantity in concentration control, while in moderation control the hydrogen content is generally specified by the $\text{H}/^{235}\text{U}$ atomic ratio. Inherent in both methods, however, is the basic assumption of a safe fuel composition, that is a $k_\infty < 1$, with adequate safeguards in effect to preclude any change in the fuel that would increase its nuclear reactivity. Thus, the following definitions of these control parameters may be stated directly:

Moderation Control. A method of assuring the nuclear safety of a system of fissionable materials whose $k_\infty < 1$, and whose $\text{H}/^{235}\text{U} < 500$,

*In the PCTR experiment a test sample is inserted in the core of an operating reactor to determine the multiplying properties of the material.

and the corollary,

Concentration Control. A method of assuring the nuclear safety of a system of fissionable materials whose $k_{\infty} < 1$, and whose $H/^{235}\text{U} > 500$.

THE EFFECT OF HYDROGEN MODERATION ON THE MAXIMUM SUBCRITICAL ^{235}U ENRICHMENT OF UF_6

Figure 3

APPLIED MODERATION CONTROL

Since very large quantities of UF_6 can be handled in a nuclearly safe manner under moderation control, the use of any particular shipping cylinder will often be determined by such considerations as the quantity of UF_6 to be shipped, the available handling facilities, the type of carrier, etc. In Table 1, three types of cylinders are listed that have been employed at diffusion plant

installations for such routine UF_6 shipments under moderation control. Although the maximum isotopic enrichments given in the table are somewhat higher than those previously specified for 2-1/2-ton cylinders,¹ Fig. 2 shows that in each case the cylinder contains only an amount of the fissile ^{235}U material that has been determined to be safe by direct criticality experiment.

Table 1
NUCLEAR SAFETY CRITERIA FOR
MODERATION CONTROLLED UF_6 CYLINDERS

Type of Cylinder ^a (ton)	UF_6 Capacity (lb)	Contained ^{235}U (kg)	Maximum ^{235}U Enrichment (wt. %)	H/ ^{235}U Atomic Ratio ^b
2-1/2	4,800	39.7	2.7	3.3
10	21,000	128.9	2.0	4.4
14	28,000	171.8	2.0	4.4

^a Minimum cylinder spacing is 1 ft edge to edge.

^b Based on minimum 99.5% UF_6 purity. All contaminants considered as hydrogen fluoride (Fig. 4).

UF_6 Withdrawal Operations

Strict moderation control procedures are in effect during UF_6 cylinder filling operations. The UF_6 is first withdrawn as a gas from an appropriate cascade unit and then condensed for liquid phase transfer to a shipping cylinder. Since hydrogen fluoride is the only possible hydrogenous contaminant in the cascade stream,* condensing operations are based on equilibrium studies of the UF_6 -HF system¹¹ to assure that the liquid UF_6 is virtually HF-free. In addition, a nonhydrogenous material is always used as the primary condenser coolant to preclude inadvertent moderation of the UF_6 in the event of equipment failure.

Because it is often uneconomical to completely evacuate the large UF_6 cylinders after each use, an allowable cylinder "heel" is established on the conservative premise that any residual material would be water. In the case of the 2-1/2-ton cylinder a 15-lb "heel" limit is specified. This amount of water would not create an unsafe condition if mixed with any amount of UF_6 at ^{235}U enrichments to 5.0%. A proportionately higher "heel" limit is used for

*Water does not exist in the presence of UF_6 due to the hydrolysis reaction:
 $\text{UF}_6 + \text{H}_2\text{O} \rightarrow \text{UO}_2\text{F}_2 + 4 \text{HF}$.

the 10- and 14-ton UF_6 cylinders. It should be noted, however, that the concept of a safe "heel" limit is applicable only for systems whose k_∞ curves exhibit maxima. The k_∞ curves at enrichments above 5% ^{235}U have points of inflection; and criticality is possible for such systems, as shown in Fig. 1, even at an $\text{H}/^{235}\text{U}$ atomic ratio of zero.

UF_6 Purity

Since UF_6 fuel supplied to the nuclear industry must meet rigid specifications set forth in the Federal Register,¹² all cylinders of UF_6 are carefully analyzed before shipment. Enriched or depleted uranium furnished as the UF_6 must consist of at least 99.5% by weight UF_6 , while the remaining 0.5% contaminant may consist of fluorocarbons, hydrogen fluoride, and certain cations. The maximum possible hydrogen moderation is determined by making the conservative assumption that all impurity is in the form of hydrogen fluoride. In Fig. 4 the $\text{H}/^{235}\text{U}$ atomic ratio is plotted as a function of purity. It will be noted that in all cases shown in Table 1 and in Fig. 4 the moderation values obtained for the 99.5% purity UF_6 lie below the maximum subcritical enrichment curve of Fig. 3, this indicating the k_∞ eigenvalue to be less than unity.

$\text{H}/^{235}\text{U}$ ATOMIC RATIO VS. UF_6 PURITY
Impurity Considered as HF

Figure 4

NEUTRON INTERACTION CONSIDERATIONS

One definite advantage in making bulk shipments of moderation controlled low-enrichment UF_6 , as defined herein, is the virtual elimination of neutron interaction considerations in the nuclear safety evaluation. It has been previously determined that the interaction potential of such moderation controlled UF_6 is negative for low enrichments,² this corresponding to a k_∞ of less than unity. Thus, an infinite array of such cylinders would be subcritical and any possible exchange of neutrons between the units may be neglected.

In order to provide indicative answers to the question of hydrogenous moderating materials that might be interspersed between UF_6 containers, a one-dimensional transport theory code⁷ was used to calculate the k_∞ of infinite arrays of spheres containing $\text{U}(4.0)\text{F}_6$ at an $\text{H}/^{235}\text{U}$ atomic ratio of 10 with a 1-in. water jacket surrounding each sphere. A 1-in. water jacket was selected since it has been determined experimentally that 2 in. is about an optimum thickness for interspersed moderators.¹³ In Fig. 5, where k_∞ is plotted vs the sphere diameter, a maximum k_∞ value of 1.21 is found for these latticed arrays, although the k_∞ of the UF_6 fuel per se is only about 0.80. It is further noted that inclusion of a 0.20-in. thickness of steel, plus the 1-in. water jacket, decreases the k_∞ maximum to about 0.94.

While the complete water flooding of interacting arrays tacitly assumed in the preceding calculations is sometimes considered as a somewhat nebulous contingency, the array moderation afforded by packaging materials of low hydrogen content is obviously not. In Table 2 are listed results of calculations indicating the effects of water interspersed homogeneously throughout interacting arrays of 17.7-in.-diam spheres of $\text{U}(4.0)\text{F}_6$, at an $\text{H}/^{235}\text{U}$ of 10. Water densities, varying from 0.05 to 1.0 g/cc, are assumed. It will be noted that the 17.7-in.-diam sphere selected for this series of calculations is the diameter of optimum reactivity found for the steel- H_2O jacket of full water density.

Table 2
 k_∞ FOR ARRAYS OF
 17.7-IN.-DIAM $\text{U}(4.0)\text{F}_6$ SPHERES
 $\text{H}/^{235}\text{U} = 10$

Sphere Jacket: 0.2" steel + 1" H_2O		Sphere Jacket: 0.2" steel + 2" H_2O		Sphere Jacket: 0.2" steel + 4" H_2O	
$\rho \text{H}_2\text{O}$ (g/cc)	k_∞	$\rho \text{H}_2\text{O}$ (g/cc)	k_∞	$\rho \text{H}_2\text{O}$ (g/cc)	k_∞
1.0	0.929	0.30	1.03	0.20	0.928
0.70	0.959	0.10	1.04	0.10	1.05
0.40	0.982	0.05	0.979	0.05	1.05
0.10	0.846				

ANISN Code⁷ used for these calculations.

K_{∞} OF ARRAYS OF UF_6 SPHERES

Figure 5

Although the data of Table 2 indicate that some reactivity increase is obtained for jackets of reduced water densities, it should be noted that in no case does the maximum k_{∞} exceed a value of about 1.05, which is less than a k_{∞} of 1.18, found for the comparable 17.7-in.-diam sphere with 1-in. H_2O jacket, at full water density. For purposes of nuclear safety considerations, therefore, it appears appropriate to assume that the maximum reactivity for a steel container, afforded by interspersed moderation of interacting arrays, is that obtained with a 1-in. water jacket at full water density, surrounding the fissile material.

Discussion

Since modern industrial nuclear safety is concerned with establishing appropriate operational procedures and equipment designs for the prevention of accidental criticality, it is recognized that an overly pessimistic approach may be highly deplorable, just as would be the most optimistic disregard of basic principles. An attenuating conservative philosophy, often used in such judgments, is reflected by the feeling that it is preferable to make upward revisions of recommended limits, based on favorable experience, than to find it necessary to impose more restrictive criteria at a later date.

Thus, in the case of the 2-1/2-ton UF_6 cylinder, since an infinite array of such cylinders with optimum 1-in. water jacket at the 2.70 ^{235}U enrichment level and $\text{H}/^{235}\text{U}$ atomic ratio of 3.3 has a k_∞ of 0.99, the cylinder by definition is considered moderation safe.* The k_∞ 's of the lower enrichment cylinders are correspondingly smaller. It should perhaps be reiterated at this point that all of the criticality data obtained for the undermoderated uranium hexafluoride system have been obtained by transport theory calculations. Thus, while there are some indications that relaxation of the moderation control limits could be made eventually, such action would probably require additional experimental checks, probably of the PCTR type, to further validate the calculations.

It may be further noted that, in using the ^{235}U enrichment limits of Table 1, consideration should be given not only the criticality aspects of moderation control, which are now reasonably well defined, but also to the important engineering and administrative problems involved in assuring that nuclear safety is maintained. For example, an extensive container test and development program has been undertaken at the Oak Ridge Gaseous Diffusion Plant to assure the proper design of containers to withstand transportation accident conditions of collision, fire, and flooding. Such containers as have now been developed¹⁴ have successfully passed rigorous tests designed to demonstrate their structural integrity. The use of such containers is thus a further guarantee that the nuclearly safe parameter of moderation control will not be invalidated.

*In this determination the cylinders were assumed to be of infinite length.

Acknowledgment

The assistance of Dr. J. R. Knight of the Oak Ridge Gaseous Diffusion Plant Central Data Processing Facility is acknowledged.

APPENDIX I

NORMALIZATION OF THE HANSEN-ROACH CROSS SECTIONS

J. R. Knight
Central Data Processing

Since it has been found that the 16-group Hansen-Roach cross sections¹⁵ appear to give lower values of k_{∞} for undermoderated uranium than has been found by ORNL and PCTR experiments,^{8,9,16} the ^{238}U absorption cross sections in groups 8 through 12, which include the resonance region, were adjusted by applying reduction factors such that the computed and experimentally determined k_{∞} values were in agreement. These reduction factors, which vary with the scattering cross section per ^{238}U atom, Σ/N_{28} , were extended to include the values that are noted below:

Σ/N_{28}	Reduction Factor	Σ/N_{28}	Reduction Factor
12	0.49	100	0.71
20	0.54	110	0.72
25	0.56	120	0.74
30	0.58	130	0.75
35	0.60	140	0.76
40	0.61	160	0.78
45	0.62	180	0.80
50	0.63	200	0.82
55	0.64	220	0.83
60	0.65	240	0.85
65	0.66	260	0.87
70	0.67	280	0.88
75	0.68	300	0.90
80	0.69	330	0.93
85	0.69	360	0.95
90	0.70	400	1.00
95	0.71		

APPENDIX II

BASIC CRITICAL MASS DATA OF FIG. 2

²³⁵ U Enrichment (wt. %)	Reflector	²³⁵ U Critical Mass (kg)	Reference
$H/^{235}\text{U} = 10$			
95.2	Paraffin	15.8 ^a 15.5 ^b	17
37.5	Water and paraffin	29.0 ^c 27.4 ^b	17
18.8	Water and paraffin	50.0 ^c 45.0 ^b	17
12.5	Water and paraffin	130.0 ^c 110.0 ^b	17
$H/^{235}\text{U} \sim 0.1$			
93.0	Water	99.6 ^d	This report
37.5	Water and paraffin	183.8 ^a 148.5 ^b	17
18.8	Water and paraffin	443.6 ^a 362.3 ^b	17

^a Value measured experimentally.

^b Experimental value reduced to spherical geometry.

^c Interpolated experimental value.

^d Computed value spherical geometry, ANISN code.

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