

**PYROPHORICITY OF URANIUM (U)**

**By**

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March 1992

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Reviewing  
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Date: 3-4-92

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# **NRTSC**

NUCLEAR REACTOR TECHNOLOGY  
AND SCIENTIFIC COMPUTATIONS

Keywords:  
pyrophoric  
ignition  
oxide coating  
temperature gradient

Retention - Permanent

## **PYROPHORICITY OF URANIUM (U)**

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**ISSUED: March 1992**

*G.C. Kao, 3-4-92*

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**Derivative Classifier**

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Westinghouse Savannah River Company  
Prepared for the U. S. Department of Energy under Contract DE-AC09-76SR00001  
Presently under Contract DE-AC09-88SR18035

**PROJECT:**

**DOCUMENT:** WSRC-TR-92-106

**TITLE:** PYROPHORICITY OF URANIUM (U)

**TASK:** 92-028-1

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## SUMMARY

Uranium metal is pyrophoric and is capable of self-ignition in air provided conditions are favorable. Based on the data in this report, spontaneous ignition of spherical particles larger than 1/16 inch in diameter would not be expected to occur in air at room temperature (25°C).

The rate at which the uranium surface oxidizes in air, balanced against the rate at which the heat of reaction is lost to the surroundings, determines whether spontaneous ignition can occur. Heat loss to the surrounding environment depends on the thermal conductivity of the uranium including the oxide coating, and on the temperature gradient.

The ignition temperature for uranium metal particles is a function of particle geometry, size or specific surface area<sup>2</sup>, heating rate<sup>3</sup>, gas composition<sup>2</sup> as well as the quantity and distribution of powder within a storage container<sup>4</sup>. The most important variable; however, affecting the ignition temperature for single samples was found by Schnizlein and Bingle<sup>4</sup> to be the specific surface area (surface area per gram) of the uranium particles.

The ignition temperatures calculated from ANL data for 1/16, 1/4, and 1/2 inch diameter spherical particles are 333, 375, and 399°C, respectively. The accuracy is believed to be about  $\pm 10\%$ , which is based on theoretical and experimental results.

## 1.0 INTRODUCTION

Fine uranium powders are pyrophoric, and under certain conditions will ignite and burn in air (oxygen)<sup>1</sup>. The self ignition of uranium powders was studied at Argonne National Laboratory (ANL) in the 1950's<sup>2,3,4,5</sup>. The data, determined from burning curves, are reported in this report to assist in the evaluation of the susceptibility of uranium metal to spontaneous ignition during storage. The literature review was done at the request of 300-Area personnel and will be used to respond to DOE order 5480.5.

## 2.0 DISCUSSION

Important parameters and data for self ignition of uranium metal particles were obtained from a literature review. Only results from experimental tests reported in the literature are discussed, and per 300-Area request, no recommendations are made for safe storage or handling of uranium metal.

### 2.1 Specific Surface Area

The ignition temperature for uranium strongly depends on the specific surface area of the metal particles. For spherical particles of uranium, the specific surface area is given by the equation

$$S_a = 3209/d \quad (1)$$

where the particle diameter is in microns and the specific surface area in cm<sup>2</sup>/g. The linear relationship relating the specific surface area and particle diameter is

$$\text{Log } S_a = - \text{Log } d + 3.50 \quad (2)$$

and is plotted in Figure 1. This relationship is based on logarithms to the base 10.

The specific surface area in equation (1) is inversely proportional to the particle diameter, so the specific surface area increases as the particle diameter decreases. A 1/16 inch (1588 micron) spherical particle is calculated to have a specific surface area of about 2 cm<sup>2</sup>/g and weigh approximately 40 mg.

## 2.2 Ignition Temperature

Burning curves developed at Argonne were used to determine the temperature for spontaneous ignition of uranium. The curves were developed by heating powders or small pieces of uranium metal in an electric furnace in oxygen to establish the temperature for ignition. The temperature difference between the furnace and sample indicated an exothermic reaction. Ignition was apparent when the slope of the burning curve was observed to change from some lesser value to nearly vertical. The ignition temperature for the uranium particles was determined from the intersection of the extension of the nearly vertical slope with that of the preignition slope as shown in Figure 2.

Ignition temperatures obtained in air are about the same as in oxygen. This is reasonable, since reactions leading to ignition are not extensive or rapid enough to generate a diffusion barrier of nitrogen between the oxygen of the air and the metal. However, in air, the rise in temperature at ignition is less pronounced, and the flash of light considerably less intense than in oxygen<sup>3</sup>.

The relationship between the specific surface area and the ignition temperature for uranium powders was determined by Schnizlein et al. These data were used to find a relationship between the specific surface area and the reciprocal of the ignition temperature. The equation developed is

$$\text{Log } S_a = 5562.628/T_i - 8.878 \quad (3)$$

where  $S_a$  is the specific surface area in  $\text{cm}^2/\text{g}$ , and  $T_i$  is the ignition temperature in degrees Kelvin.

A plot of the Argonne data is shown in Figure 3. From the figure, particles larger than 1588 microns have an ignition temperature greater than  $333^\circ\text{C}$ . Using equation (3), the ignition temperatures for 1/16, 1/4, and 1/2 inch equivalent diameter particles were calculated to be 333, 375, and  $399^\circ\text{C}$ , respectively.

Powders of less than about 44 microns in equivalent diameter are classified as sub-sieve particles. Sub-sieve spherical powders tested at ANL had median diameters of 12.5, 20, 33, and 37 microns, respectively. These sub-sieve fractions yielded ignition



temperatures (shown in Figure 3) approximately 15 to 25 degrees C higher than values expected from extrapolation of the line obtained from sieved fractions. One explanation given for higher ignition temperatures by the experimenter was that the powders may have been partially oxidized during the separation procedure<sup>4</sup>.

Tests were also carried out by Tetenbaum et al.<sup>4</sup> using sub-sieve irregular powders. Irregular powders are expected to have a higher specific surface area than spherical ones. Each size distribution that was tested contained median diameter powders of either 6.2, 15.8, 22.5, or 28.0 microns. The four samples gave experimental ignition temperature values of approximately 150°C while extrapolation of the data in Figure 3 (not shown) or calculation of the ignition temperatures using equation (3) with an experimentally measured surface area of 10,500 cm<sup>2</sup>/g predicted an ignition temperature of about 160°C. The difference in the experimental and calculated values was about 7% for fine irregular particles.

The experiments using spherical and irregular sub-sieve powders clearly show the effect of particle geometry and specific surface area on the ignition temperature of uranium powders. The larger the equivalent spherical diameter of the uranium particle the less likely self ignition will occur at room temperature.

Murray, Buddery and Taylor<sup>6</sup> theoretically calculated the ignition temperature for uranium powders as a function of particle radius (diameter). For a 200 micron diameter particle, the theoretical value for the spontaneous ignition temperature was calculated to be 307°C. Using the ANL empirical equation (3), the ignition temperature is determined to be 280°C or approximately 9% lower than the theoretical value for small diameter particles.

### 2.3 Preoxidation

Massive uranium reacts in air at room temperature to form a yellowish appearance. After several days, if the atmospheric humidity is normal, the film will become brown-black or green-black.

A freshly fractured surface is ordinarily more reactive than an as cast surface probably because of the concentration of corrosion retarding impurities at the surfaces of castings.

A study of the effects of preoxidation on the ignition temperature of uranium powder prior to burning curve ignition was done by Tetenbaum<sup>3</sup>. Powders of 140/+170 mesh were heated at about 150°C in air for varying lengths of time. Microscopic examination of the powders revealed that the surface of the particles eventually became coated with black oxide nodules, the extent of coverage varying with heating time. Complete coverage of particles with nodules occurred after approximately fifteen hours of heating.

Experimental data showed the ignition temperature decreasing, leveling off, and finally increasing with extent of preoxidation. Spherical uranium powder between 88 and 105 microns (140/+170 US Sieve Series) exhibited the highest ignition temperature of 260°C when particles displayed some interference colors indicating slight oxidation. Partial coverage by oxide nodules (after about 8 hours at 150°C) caused a decrease in the ignition temperature to a minimum value of 235°C. With complete nodule coverage, the ignition temperature increased to 250°C.

#### **2.4 Cover Gas Effects On Ignition Temperature Of Uranium**

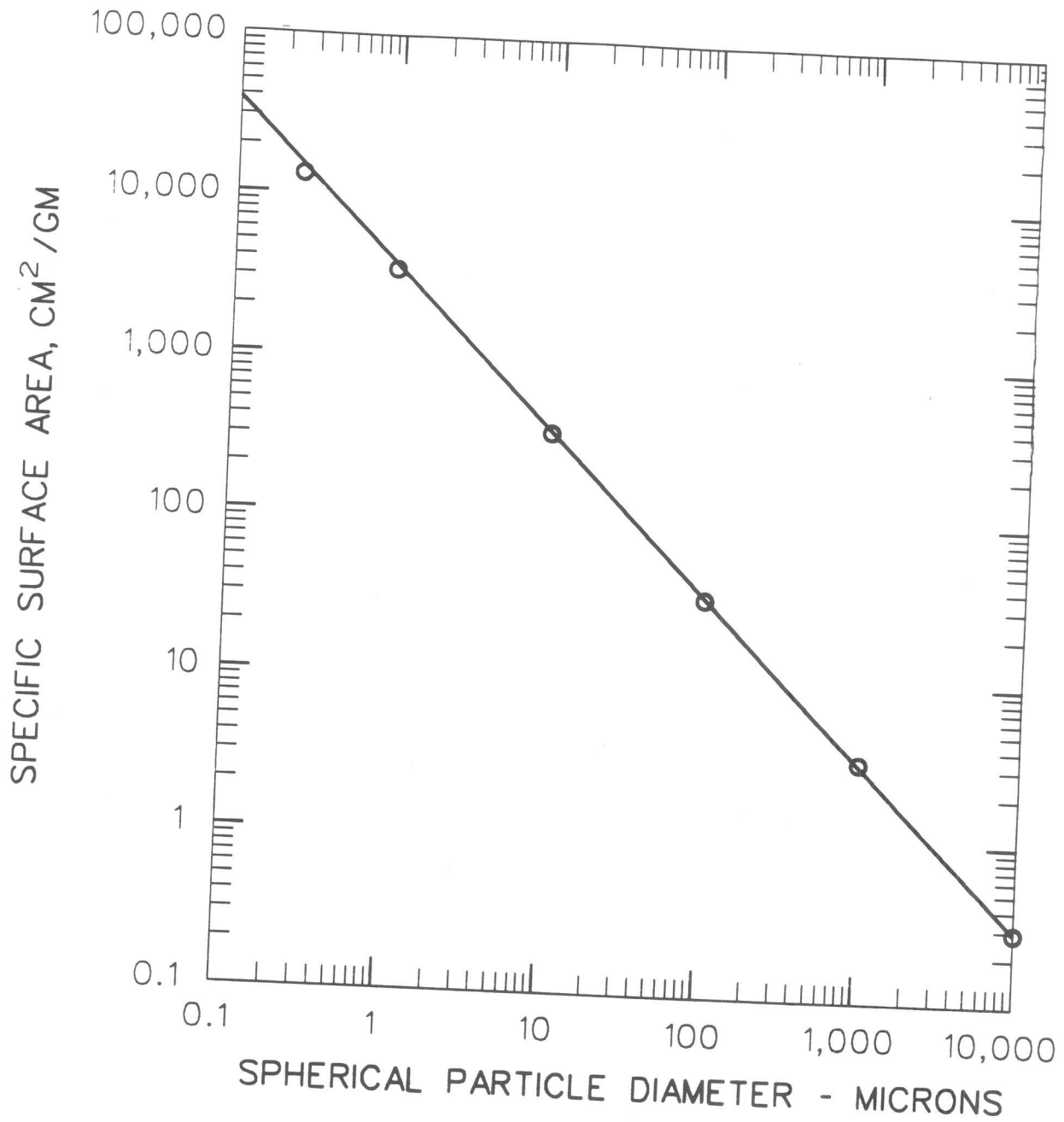
Burning curve tests were made by Schnizlein et al.<sup>2</sup> to determine the effect of gas composition on the self ignition of uranium metal. The test used 8.5 mm cubes of uranium in gas mixtures of helium and oxygen.

It was observed that a 33 per cent helium/oxygen mixture did not change the ignition temperature appreciably, but that the reaction proceeded more slowly. Concentrations of helium from 33 to 60 per cent caused a uniform increase of the ignition temperature, from 615 to 955°C. This amounted to 10 to 15 degrees in the ignition temperature per helium per cent. No ignition was observed in 80 per cent helium. It was suggested that uranium might be subject to ignition in 80 per cent helium if the sample was heated over 1000°C. Uranium does not corrode in pure, dry helium, but uranium stored for several months in static helium of normal purity may develop corrosion products.

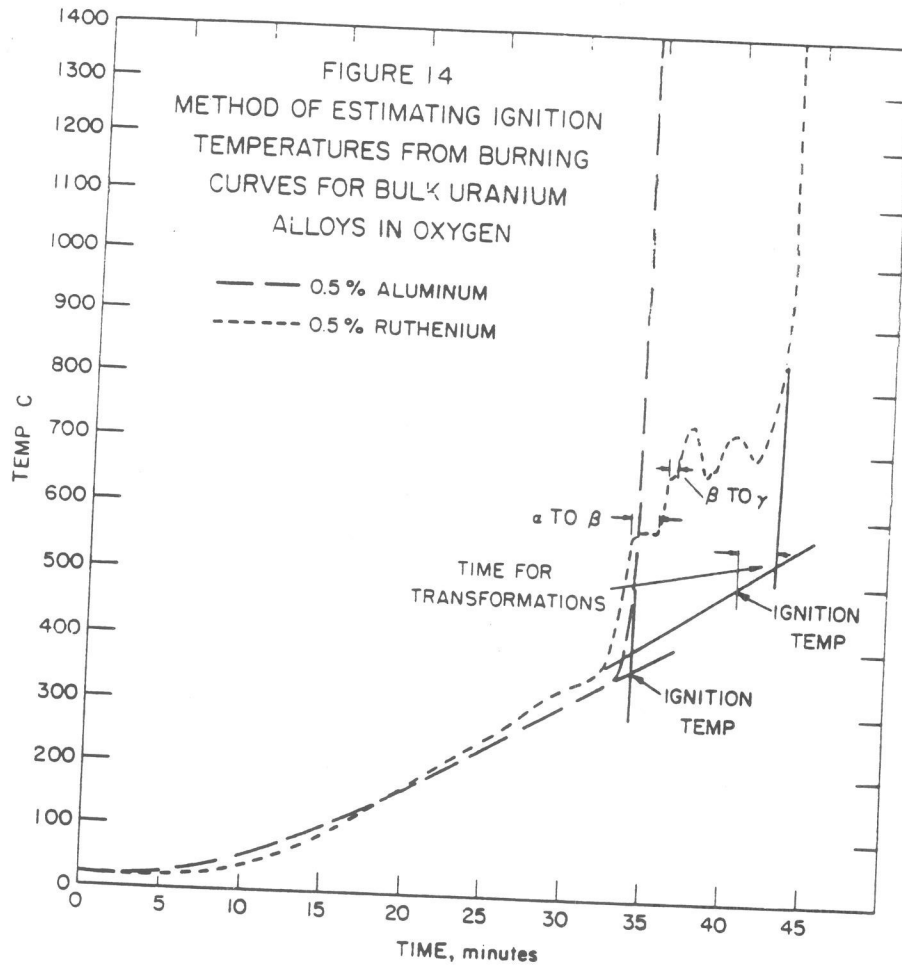
Burning depends on oxidation reactions so that elimination of all oxygen will prevent ignition provided the cover gas does not react with uranium metal. Inert cover gases must be dry because moisture will react with uranium.

### 3.0 REFERENCES

1. Uranium Metallurgy, Vol 2, Corrosion and Alloys, Interscience Publishers, A Division of John Wiley and Sons, pp. 801-815, 1962.
2. Chemical Engineering Division Summary Report, Argonne National Laboratory, July, August, and September, pp. 55-65, ANL5789, 1957.
3. Chemical Engineering Division Summary Report, Argonne National Laboratory, January, February, March, pp. 54-59, ANL5858, 1958.
4. Chemical Engineering Division Summary Report, Argonne National Laboratory, October, November, December, pp. 170-178, ANL6287, 1960.
5. Chemical Engineering Summary Report, Argonne National Laboratory, July, August, September, pp. 138-152, ANL6231, 1960.
6. Murray, P., Buddery J.H., and Taylor, J.F., "The Nature of Metal Powders," AEREM/R1428, April 27, 1954.



**Figure 1**  
**Specific Surface Area for Spherical Particles**



**Figure 2**  
**Burning Curve for Uranium**

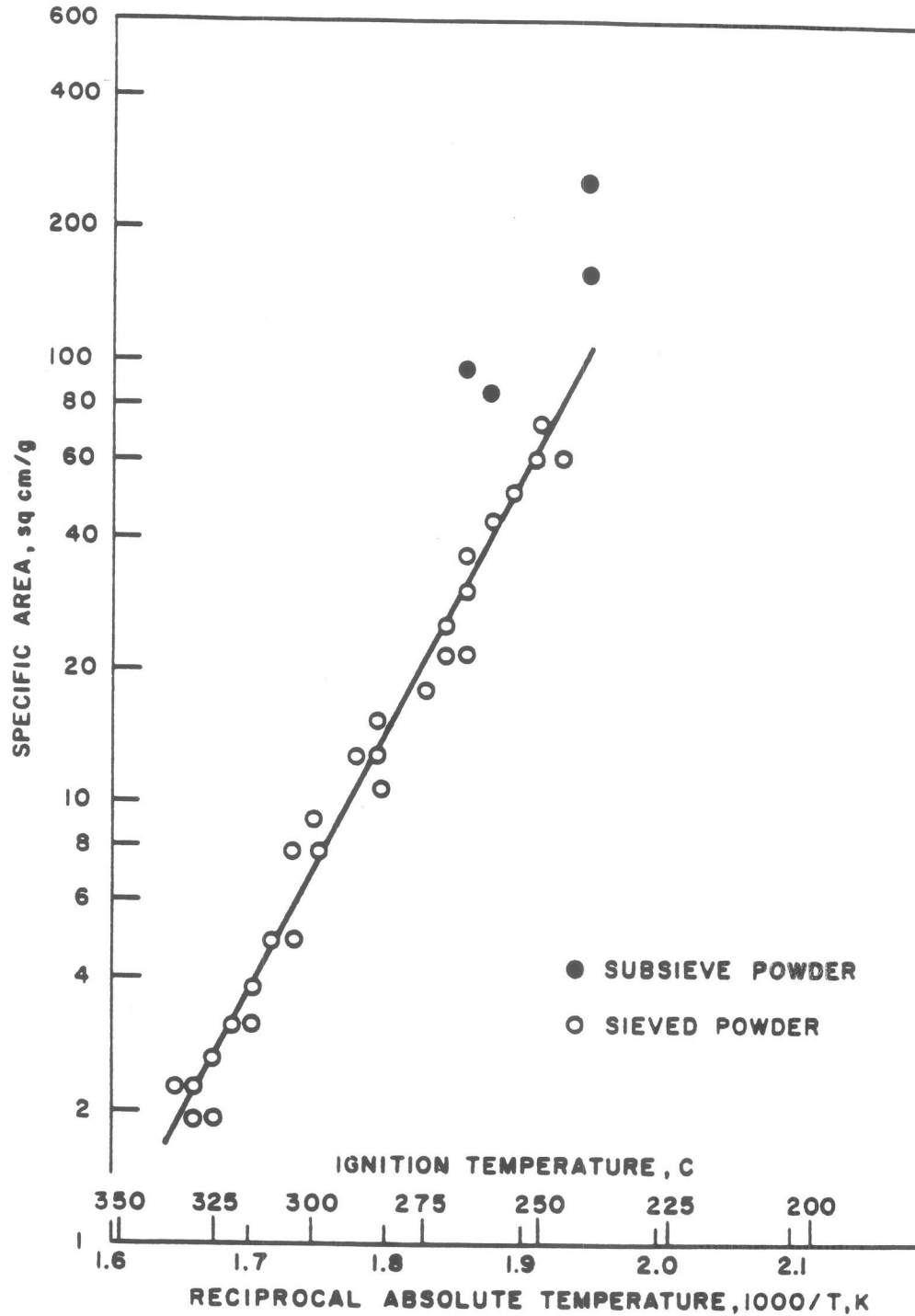


Figure 3

Effect of Specific Surface Area for Spherical Uranium  
Powders on Experimental Ignition Temperatures

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